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Demonstration of longitudinally polarized optical needles

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Abstract: Longitudinally polarized optical needles are beams that exhibit ultra-long depth of field, subwavelength transverse confinement, and polarization oriented along the longitudinal direction. Although several techniques have been proposed to generate such needles, their scarce experimental observations have been indirect and incomplete. Here, we demonstrate the creation and full three-dimensional verification of a longitudinally polarized optical needle. This needle is produced by generating a radially polarized Bessel-Gauss beam at the focus of a high numerical aperture microscope objective. Using three-dimensional spatial mapping of second-harmonic generation from a single vertically aligned GaAs nanowire, we directly verify such a longitudinally polarized optical needle's properties, which are formed at the focus. The needle exhibits a dominant polarization, which is oriented along the longitudinal direction, an ultra-long depth of field (30 λ), and high spatial homogeneity. These are in agreement with corresponding focal field calculations that use vector diffraction theory. Our findings open new opportunities for manipulation and utilization of longitudinally polarized optical needles.

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1. Introduction

Vector Bessel beams represent the non-diffractive solutions to the full vectorial Helmholtz wave equation [1,2]. They constitute the extension of the well-known Bessel beams, obtained from the scalar wave equation [3–5], to optical beams with various states of polarizations, like radially- (RP) or azimuthally-polarized (AP) beams. In principle, vector Bessel beams show an infinite depth of field similar to the well-known case of scalar Bessel beams. However, due to limitations in the physical size of utilizable apertures and available input energy, one can only create approximations of such beams in the laboratory, which are known as quasi-Bessel beams or Bessel-Gauss (BG) beams. Several methods exist to generate vector BG beams including spatial light modulators [6,7], axicons [8,9], or tuning the output mode of a laser [10]. The vectorial behavior of such BG beams is particularly important under tight focusing conditions, where longitudinal electric field components naturally arise [11–18].

Longitudinally-polarized optical needle is a term used to represent longitudinal electric fields that show spot-like intensity distribution in the transverse plane with sub-wavelength confinement over a distance of several λ along the longitudinal axis (i.e., beam propagation axis), where λ is the excitation wavelength. Such optical fields are expected to be valuable in providing alternative illumination conditions in an optical microscope [19–21], increasing optical data storage density [22] or multi-plane micromanipulation of nanoparticles [23,24]. Various methods, based on RP beams, have theoretically predicted the generation of optical needles using diffractive optical elements consisting of several belts [25–29] to promote constructive and destructive interferences at the focal region. The tight focusing of RPBG

beams has been shown to be a practical way to generate longitudinally-polarized optical needles [30–34]. To date, however, experimental verifications of the generation of longitudinally-polarized optical needles have been limited and mostly relied on the use of indirect measurement techniques [30,31,34,35] thereby substantially hindering the full characterization and subsequent tailoring of such needles in three dimensions.

Here, we experimentally demonstrate the creation and verification of a longitudinallypolarized optical needle generated by an RPBG beam at the focus of a high numerical aperture (NA) microscope objective. By forming a thin annular RP beam at the back focal plane of the microscope objective using polarization mode converter and an axicon in tandem, we create an RPBG beam at the focus of the microscope objective. We then probe directly the properties of the longitudinal electric field of such a beam by spatially mapping the secondharmonic generation (SHG) from a single vertically-aligned GaAs nanowire excited with the RPBG beam. By raster-scanning such nanowires, known to be mainly responsive to longitudinal electric fields [36–40], the three-dimensional spatial distribution of the optical needle is obtained unambiguously. The results are then compared with the SHG measurements from the same nanowire using RP and AP beams and an azimuthally-polarized Bessel-Gauss (APBG) beam. The experimental results are in good agreement with focal field calculations using vectorial diffraction theory.

2. Methods

2.1 Optical microscopy setup

The optical microscopy setup used to generate the vector beams and to probe the resulting optical needles at the focal region is shown in Fig. 1. The excitation beam originated from a mode-locked laser (excitation wavelength of 1060 nm, pulse length of 140 fs, repetition rate of 80 MHz). First, we modified the excitation path of our custom-built cylindrical-vectorbeam-equipped point-scanning nonlinear optical microscope [36–39,41,42] to incorporate the generation of the RPBG and related vector beams. This is done by converting the linearlypolarized Gaussian output of the laser into an RP or AP doughnut-shaped beam using a commercial polarization mode converter (MC, Arcoptix, S.A.). We then used this beam as input to an axicon (apex angle of 176°) to generate a vector BG beam [8]. In order to generate a similar beam at the focus of the microscope objective, we focused our vector BG using a set of relay lenses (RFL) to generate a thin annular RP or AP beam with the proper size (around 6 mm in diameter) at the back focal plane of the microscope objective (MO) itself [15]. The resulting thin annular RP and AP beams, located at the back focal plane of the microscope objective, were imaged with a beam profiler. Profiles were acquired by placing a beam splitter before the microscope objective to direct part of the beam to the beam profiler placed at the correct distance.



Fig. 1. Schematic diagram of the optical microscopy setup. HWP1/2: half-wave plate, PBS: polarizing beamsplitter, BD: beam dump, M: mirror, SF: spatial filtering, MC: mode converter, BE: beam expander, RFL: relay formation lenses, DF: dichroic filter, MO: microscope objective (NA = 0.8, $50 \times$), PZ: piezo-stage scanner, S: sample mounted on a glass slide, F: filter, PMT: photo-multiplier tube. The full red line represents the path of the fundamental laser beam; the dashed turquoise line represents the path of the SHG signals collected in epidetection.

To confirm the polarization purity of the vector BG beams before focusing, we acquired the thin annular RP and AP beams at different analyzer settings, and are shown in Fig. 2. After creating the appropriate laser beam, the latter was directed to a high NA (0.8) objective to focus the beam onto our sample that is mounted on a computer-controlled three-dimensional piezo stage-scanner (PZ). In all our measurements, the sample is attached to a glass slide (S). We then collected the total SHG originating from our sample in epi-detection through the same microscope objective. The excitation and SHG wavelengths were discriminated using appropriate dichroic (DF) and bandpass filters (F) and the SHG signal was detected using a photo-multiplier tube (PMT).



Fig. 2. Experimental beam profiles of the generated thin annular RPBG and APBG beams. The beams appear at the back focal plane of the microscope objective. The beam profiles were recorded using a beam profiler without and with an analyzer at different orientations (see double-headed yellow arrows).

2.2 Sample

Our sample is composed of vertically-aligned zinc-blende GaAs nanowires grown on a GaAs substrate using selective-area metallo-organic vapour phase epitaxy [37,43]. For this

experiment, these nanowires are arranged in a periodic lattice with a pitch of 2.5 μ m, have a diameter of 90 nm and a length of about 2.6 μ m. We have demonstrated in previous works that SHG from such nanowires is driven by the longitudinal electric field and using point-scanning allows us to therefore map the longitudinal field distribution of a focused vector beam in three dimensions [36–40]. The separation between each nanowire is large enough to avoid overlap of SHG signals and related SHG verification measurements (i.e., spectral and power dependence) were performed in previous works [36,37].

2.3 3D SHG microscopy parameters

In order to evaluate the three dimensional character of the generated optical needle, we performed systematic transverse (xy) and longitudinal (xz or yz) raster scans of the GaAs nanowire sample at the focal region. It is also important to note that, independently of the input polarization, we always compared SHG signals for the same GaAs nanowires. Transverse scans were performed over a $10 \times 10 \text{ }\mu\text{m}^2$ area and longitudinal scans over a 7 × 15 µm² area for the data sets in Figs. 3 and 4. In Fig. 5, we used the maximum longitudinal scanning range of our system and scanned nanowires over a $7 \times 50 \ \mu\text{m}^2$ area. In all scanning measurements, a pixel resolution of 66.7 nm was used and maintained for each Cartesian axis. All scanning, i.e., pixel-by-pixel, maps were recorded using an average input power of 2 mW and a pixel-dwell time of 50 ms. Prior experiments showed that these exposure conditions are well below the damage thresholds of the used sample. All SHG intensity maps were plotted directly from the raw SHG counts recorded by the PMT using MATLAB, i.e., without image processing or related reconstruction. Each of these maps was independently normalized according to the minimum (Min) and maximum (Max) count values recorded for each scan. All maps also show a multiplication coefficient applied to all the pixel values in one image to show the relative strengths of the SHG signal levels with respect to the reference.

2.4 Focal field calculations

The focal field distributions were calculated using the formalism described by Richards and Wolf [11] for tight focusing of optical fields. Figure 6 shows the calculated transverse (E_T) and longitudinal (E_Z) electric field distributions in xy (z = 0) and xz (y = 0) planes for the different input vector beams and for settings that closely simulate our experimental conditions (i.e., excitation wavelength of 1060 nm,a focusing angle of 45°, the latter representing a NA below 0.8 due to under filling conditions in our experiments, environment of air). The angle between rays focused by the objective is taken to be 3° for the case of RPBG and APBG beams.

3. Results

We used 3D SHG microscopy of GaAs nanowires (See Methods) to verify the longitudinal electric fields at the focus of the microscope objective. We demonstrated in previous works that the SHG signal from a single vertically-aligned nanowire originates mainly from nanowire and not from the substrate, and showed that such nanowire is responsive mostly to longitudinal electric fields [36,37]. In addition to this, and considering the rather short length of our nanowires ($2.6 \mu m$), by performing 3D SHG microscopy scans in a plane containing a nanowire and parallel to the longitudinal direction, one can get a good approximation of the depth of field of the longitudinal field created by a focused RP beam [40]. For reference, we first used an RP beam at the input of the microscope objective to excite the nanowires. This was done by removing the axicon and relay-formation lenses (RFL) after the mode converter (MC) in the optical setup displayed in Fig. 1. As seen in Fig. 3(a), a SHG transverse raster scanning map reveals several point-like distributions that are centered with respect to the location of each GaAs nanowire. This is expected considering that the longitudinal electric field of a focused RP doughnut beam shows such a point-like distribution [13] when viewed in the transverse plane. This characteristic transverse distribution is also consistent with our

previous works [36,37]. The average count value from each nanowire (the substrate) is about 100 000 per 50 ms (2200 per 50 ms) in Fig. 3(a). We then acquired a longitudinal scanning map in a plane containing three arbitrarily chosen nanowires. The dashed yellow line in Fig. 3(a) shows the y coordinate along which we scanned these three nanowires in the xz plane. Figure 3(b) shows the corresponding longitudinal SHG intensity map from these three nanowires. From this scanning map, we clearly distinguish the location of the nanowires and can estimate the distance over which they were excited using the RP beam. The SHG counts recorded by the photo-multiplier tube (PMT) for this longitudinal map are similar to the ones recorded for the transverse scan for the brightest nanowire. Some variations in SHG signals from one nanowire to another can be seen in the longitudinal scan and are due to slight misalignment of the sample plane with respect to the scanning axes as seen in the transverse scan of Fig. 3(a). Such variations can also be observed in other longitudinal scans. From the SHG intensity maps of Figs. 3a and 3b), we calculated the full width at half maximum (FWHM) value to estimate the transverse resolution. From both scans, we retrieved a consistent FWHM value of 0.42 λ , the reference being the excitation wavelength λ . We also calculated the FWHM over the longitudinal direction from Fig. 3(b) and obtained a value of 2.2λ

We then implemented the necessary optical components in our microscopy setup to generate an RPBG and acquired similar SHG measurements from the nanowires in the same regions of interest to determine whether we were able to create a sub-wavelength longitudinally-polarized optical field with extended depth of field at the focus of the objective. The input beam used for this was an RP ring in order to generate a tightly focused RPBG beam at the focus of the objective. The SHG signals from the GaAs nanowires of Fig. 3(c) show again a point-like distribution with a non-negligible surrounding ring, revealing the transverse spatial distribution of longitudinal electric field of an RPBG at the focus of a high NA objective. This result confirms earlier simulations as well as indirect observations of such longitudinal electric field distributions [34,44,45]. The average SHG count value from each nanowire (the substrate) is about 6000 per 50 ms (110 per 50 ms) in Fig. 3(c), which is significantly lower compared to the count level obtained using an RP doughnut beam, this mostly due to the extended depth of field observed using an RPBG beam. The surrounding ring shows a count value of around 350 per 50 ms.



Min

Max

• 1µm

×23

d)

Min

Max

<u>1um</u>

C)

zĊ



×22 🧧

Fig. 3. 3D SHG intensity maps acquired from GaAs nanowires excited by the respective vector beams. (a,c) Transverse and (b,d) longitudinal scanning maps of GaAs nanowires using (a,b) RP and (c,d) RPBG beams. The dashed yellow lines in the transverse scanning maps represents the plane along which the longitudinal scans were performed. The relative SHG signal levels for these data sets are shown.

Similarly, we performed a longitudinal scan of the same nanowires of interest (see marks in Fig. 3(c)) using the RPBG beam. As seen in Fig. 3(d), the SHG signal coming from the three nanowires is almost constant over the 15 μ m range. This result confirms that the focused beam used seems to exhibit an almost constant, i.e., homogeneous, longitudinal electric field distribution along the optical axis over a long range, while maintaining a spot-like transverse distribution when viewed in the transverse plane. We calculated the transverse FWHM value from Fig. 3(c) and 3(d) and retrieved in both cases a value of 0.41 λ , close to the value obtained for the RP doughnut beam. However, when we looked at the FWHM over the longitudinal range using Fig. 3(d), the value obtained was 11.4 λ , confirming the creation of a longitudinally-polarized optical needle.

To confirm the longitudinal polarization of the optical needle created, we performed the same set of experiments as in Fig. 3, but this time using an AP and APBG beam. This was done by simply switching from RP to AP beam using built-in electronics of the commercial mode converter (MC). Using the axicon after the AP beam then allowed us to generate an APBG beam. In this experiment, we intended to prove that the contrast observed in Fig. 3 using the RP beam and the RPBG beam originates dominantly from longitudinal electric fields. Focused AP and APBG beams, do not exhibit any longitudinal electric field in the focal volume [13,45]. Therefore, only, i.e., purely, transverse electric fields are created at focus of such beams. Prior to scanning, we ensured that switching the incident polarizations, i.e., from RP(BG) to AP(BG), did not affect the beam position at focus.

Figure 4 shows the SHG intensity maps obtained for the transverse and longitudinal scans for both AP and APBG beams. In Fig. 4(a), the transverse scan using an AP beam shows low SHG signals, about 100 times lower compared to the one obtained using an RP beam. More



importantly, even though some features can be distinguished, SHG counts from the substrate (1300 per 50 ms) are close to the maximum SHG counts of the features observed (1600 per 50 ms). The lowest SHG counts are about 800 per 50 ms. We also performed a longitudinal scan, displayed in Fig. 4(b), where the substrate is clearly seen, with SHG levels similar to the transverse scan. The same set of scans was then acquired using an APBG beam and are displayed in Figs. 4c) and 2d). SHG counts are even lower than for the AP beam, with levels around 40 per 50 ms. No features appear in the transverse scan and no contrast emerges from the nanowires. This clearly suggests that the nanowires respond mostly to longitudinal electric fields and that the focused beam, in both cases, contains mostly transverse electric field components. It also confirms that the SHG signals collected using an RPBG beam emerged from the presence of strong longitudinal field along the nanowires.



Fig. 4. 3D SHG intensity maps acquired using GaAs nanowires excited by the respective vector beams. (a,c) Transverse and (b,d) longitudinal scanning maps of GaAs nanowires using AP doughnut and APBG beams. The dashed yellow line represents the plane along which the longitudinal scans were performed. The relative SHG signal levels for these data sets are shown, with Fig. 3 as reference.

The results of Fig. 3(d) show that the longitudinal electric field is almost homogeneous at least over the initial range of 15 μ m. In order to fully evaluate the extent of the optical needle created at, we performed new longitudinal scans using RPBG and APBG beams but this time over the full available range of our piezo-scanning system (PZ), this range being 50 μ m. Results from these scans appear in the SHG intensity maps of Fig. 5. Figure 5(a) shows that the contrast is preserved over the full 50 μ m range with a maximum count level at 7500 per 50 ms when using the RPBG beam. Some inhomogeneities can be seen along the needle, and the maximum count level occurs in the more intense SHG signal found at the beginning of the needle. The central part shows count levels around 6500 per 50 ms. The minimum count level between the beginning and central part of the needle 3500 per 50 ms. The transverse FWHM



value is also preserved when compared to Figs. 3(c) and 3(d). The SHG signals, which are closely linked to the longitudinal electric field at focus, extend over 40 to 50 μ m. It is, however, difficult to calculate the longitudinal FWHM considering the few small variations seen along the needle. We believe that a good approximation is around 30 λ . In addition, Fig. 5(b) shows the results of a longitudinal scan using the APBG beam. As expected, no contrast emerges from the nanowires and SHG count levels remain low.



Fig. 5. Comparison of SHG maps acquired using respective vector beams. Longitudinal scanning maps of three nanowires over a 7×50 µm range of the piezo-stage scanner, using (a) RPBG and (b) APBG beams. The relative SHG signal levels for these data sets are shown, with Fig. 3 as reference.

To support our experimental results, we performed simulations of the focal fields using RP and AP beams and their thin annular beam counterparts as input before the objective (See

Methods). These simulations confirm the experimental results obtained in Figs. 3, 4, and 5. The longitudinally-polarized optical needle can be particularly seen in Fig. 6(b). It confirms the longitudinal FWHM of the RPBG beam evaluated in Fig. 5, which is also about 30 λ when evaluated from Fig. 6(b). The transverse FWHM of the RPBG beam, also evaluated from Fig. 6(b), is about 0.84 λ at the wavelength of 1060 nm, in good agreement with the 0.41 λ transverse FWHM calculated from SHG signals of Fig. 3(b). Finally, Fig. 6(d) confirms that an APBG does not show any longitudinal electric field at focus.



Fig. 6. Calculated focal fields of the RP, RPBG, AP and APBG vector beams. Focal field distributions of a tightly focused a) RP beam, b) RPBG beam, c) AP beam, and d) APBG beam in the (x,y,z = 0) and (x,y = 0,z) planes for an excitation wavelength of 1060 nm and for a focusing angle of 45° in air. $|E_T|$ represents the transverse electric field component and $|E_Z|$ represents the longitudinal electric field component. Both longitudinal (x,y = 0,z) and transverse (x,y,z = 0) planes are shown. Simulations were run over a 5.5 × 50 µm area for the longitudinal plane and over a 2 × 2 µm2 for the transverse plane. The relative electric field amplitudes between $|E_T|$ and $|E_Z|$ are shown for each vector beam.

For completeness, we also probed the self-healing property of RPBG beams [7,46,47] and the resulting longitudinally-polarized optical needles generated at the focus of the microscope objective. After blocking a quarter, and then a half of the RPBG beam after the axicon with an opaque metallic corner, and represented in Fig. 7, we performed the same SHG longitudinal scans described before over a $7 \times 15 \,\mu\text{m}^2$ area. The results revealed that the SHG signals remain homogeneous, confirming the self-healing property of an RPBG beam. The input power was not compensated for the presence of the block and therefore SHG signals show lower counts, however consistent with the amount of input power blocked.



Fig. 7. Robustness of the longitudinally-polarized optical needles created at the focal region. Longitudinal SHG intensity maps using three nanowires excited by the quarter- or half-blocked RPBG beam. The scanning maps cover a $7 \times 15 \,\mu m$ area. The relative SHG signal levels for these data sets are shown, with Fig. 3 as reference.

4. Discussion

The above results show that we managed to experimentally create and directly probe the longitudinal electric field generated by an RPBG beam at the focus of a microscope objective using three-dimensional mapping of the SHG from single GaAs nanowires. The SHG longitudinal scans suggest that the longitudinally-polarized optical needle extends over 30 λ in the longitudinal direction and shows a transverse spot-like distribution with sub-wavelength confinement of 0.41 λ .

Comparing SHG signal levels in Fig. 3 suggests that the RPBG beam provides a significantly lower SHG response from an individual nanowire than the RP doughnut beam, for the same input power of 2 mW. This can be explained by comparing Fig. 3(b) to Fig. 3(d), and even Fig. 5(a), which reveal the depth of field of the longitudinal field of each beam at focus. The depth of field of the RPBG at focus is seen to be almost 20 times larger than the one of an RP doughnut beam. The energy is thus spread over a larger distance, which explains the lower SHG response when an RPBG beam is used. Moreover, a non-negligible portion of the longitudinal electric field component is contained in the characteristic surrounding ring of the RPBG beam, which further explains the lower SHG signal coming from a single nanowire. This is also put in relation to the 2.6 μ m length of the nanowires used in the experiments, therefore best excited with an RP doughnut that provides a 2.2 λ long longitudinal electric field at focus. However, an RPBG beam could be a better choice to excite longer nanowires, or other elongated nanostructures sensitive to longitudinal electric fields. Moreover, such a longitudinally-polarized optical needle could bring even better sensitivity for three-dimensional imaging of extended nanostructures [19,21,40,48].

Figure 5(a) shows that the optical needle created at focus is not perfect and exhibits some inhomogeneities along the longitudinal direction. Such artifacts can be explained mainly by the high sensitivity of the BG beam to the axicon tip sharpness [49,50]. An axicon tip parameter with roundness above 10 μ m is enough to generate strong oscillations [49], explained by destructive interferences at focus. However, it is important to remember that in our nonlinear measurements, the SHG scattered signals were collected and not the linearly scattered signals. Therefore, the collected SHG intensities and the actual longitudinal electric field of the fundamental beam at focus are linked via the relation $I(2\omega) = E^4(\omega)$. For instance, the difference in SHG intensity between the two maxima of the needle (lower and central portion of maps in Fig. 4(a)) is below 20%. This corresponds to a difference of 5% in terms of electric field at the fundamental frequency. The differences in terms of SHG intensity between the maximum and minimum variations along the needle are contained below 35%, which corresponds to a difference of 10% in terms of electric field at the fundamental frequency. Overall, even though the SHG longitudinal scans show some imperfections, the optical needle possesses a great degree of homogeneity at the fundamental frequency ω .

The GaAs nanowires used in these experiments being sensitive only to longitudinal electric fields, it is hard to quantify experimentally the possible remaining proportion of transverse electric field at focus. However, considering the spatial distribution of the transverse component of an RPBG [1,45], and observable in Fig. 6(b), we can state that the optical needle created at focus is predominantly longitudinally-polarized. In more detail, based on our experimental conditions and according to Table 2 of [15], we can expect the longitudinal electric field component of the total beam at focus to be between 3 to 10 times higher than its transverse electric field only twice greater than the transverse electric field component. If the goal is to enhance even more the proportion of electric field polarized along the longitudinal direction, one can tune parameters such as the divergence angle or the focusing angle.

The physical properties of the nanowire influence the effective imaging performance of our SHG microscope. In our case, both height and diameter of the semiconductor nanowire need to be considered. In fact, we found before that the smallest nanowire probe that can be used to map the longitudinal electric fields via SHG exhibits a length of 100 nm and a diameter of 40 nm [36]. The level of SHG signals acquired using this nanowire, however, is relatively low. It is also not practical to use very long nanowires to map the 3D spatial distribution of longitudinal electric fields. This is because in long nanowires, e.g., about 10 μ m (assuming that the diameter is 40 nm), the electromagnetic modes aside from the inherent structure of the nonlinear tensor could also strongly influence the SHG efficiency. To date, the role of these electromagnetic modes in the SHG efficiency of the nanowire remains unknown.

For future work, it is valuable to explore the possibility of evaluating the 3D polarization purity of the optical needle. This task is possible but this will demand the development of new techniques and materials used as probes. It is important to remember that the full experimental measurement of all polarization components of an optical needle is complicated and not available to date. This is because both in- and out-of-plane electric fields of the optical needle should be measured directly at high reliability. So far, the probes used in farfield optical scanning microscopy to directly evaluate the focal fields are either sensitive to the total electric field or a particular electric field component only. Note that the implementation of full polarization (including amplitude and phase) measurements, which are based on either optical field reconstruction [51] or near-field nanoprobes, is not easy in practice due to the large spatial extent of the optical needle.

In conclusion, we have demonstrated the experimental creation of a longitudinallypolarized optical needle created by an RPBG beam at the focus of a high NA microscope objective. The needle was probed directly using three-dimensional SHG microscopy of single

and vertically-aligned GaAs nanowires. We believe that the ability to generate and verify longitudinally-polarized optical needles will be useful for manipulating several nanoparticles on one line, optical microscopy, and optical data storage.

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