3D nano surface profilometry by combining the photonic nanojet with interferometry

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Abstract. Interference microscopy has become a standard technique for measuring microscopic surface roughness, being rapid, non-contact and having a high nanometric axial resolution. Nonetheless, a limitation is the lateral resolution of $\lambda/2$ due to diffraction. Amongst the many recently developed techniques for improving the lateral resolution in unlabelled far field imaging, the use of microspheres placed on the sample has recently been extended from 2D imaging to 3D measurement by combining it with interferometry. In the present work, results are shown of combining the high lateral resolution of the photonic nanojet produced by the microsphere with interference microscopy to achieve sub-diffraction limited lateral resolution with nanometric axial resolution. This is achieved by imaging through a microsphere placed on top of the sample in front of the interference objective in a Linnik setup. Results of the 3D measurements of narrow gratings through the microsphere are presented, that are not observable with the same objective in air since they are below the resolution limit. Simulations of the interaction between the photonic jet and the grating leads to a basic understanding of the image formation. This new technique opens new possibilities for high resolution characterization in nanomaterials and the biological sciences.

1. Introduction
Interference microscopy has become a standard technique for measuring microscopic surface roughness and 3D surface structures [1]. While it has the advantages of having a very high nanometric axial resolution and being rapid since it operates using classical imaging in the far field, it suffers from the limits of diffraction, resulting in a lateral resolution of $\lambda/2$, which is over two orders of magnitude worse than the axial resolution. This can be calculated from the criterion of $R_c = K\lambda/NA$, where $R_c$ is the distance between two point sources, $NA$ is the numerical aperture, with $NA = n\sin\theta$ where $\theta$ is the half angle of the light cone that can enter the lens (objective) from a point source, $n$ is the refractive index of the object-space and $K$ is 0.61 or 0.47 depending on whether the resolution definition comes from the Rayleigh criterion or Sparrow criterion respectively [2]. By decreasing the wavelength and increasing the NA, at best the lateral resolution can be improved to a value of 200 nm [3]. Near field techniques have clearly overcome the lateral resolution problem, but remain limited by the need for point scanning [4].

In far field optical microscopy, several techniques have been proposed to improve the lateral resolution (see classification scheme in [5] for example). Notably, stimulated emission depletion (STED) microscopy [6], photoactivated localization microscopy (PALM) [7] and stochastic optical
reconstruction microscopy (STORM) [8] have brought the field of optical nanoscopy to the forefront with the Nobel Prize for Chemistry of 2014 [9]. While these techniques result in a lateral resolution of several tens of nm, they nonetheless require labelling with fluorescent dyes or high light intensities, both of which can be destructive to living matter. In addition, markers cannot easily be used in materials characterization.

Some of the main principles of label-free far field nanoscopy for materials characterization were studied at the beginning of the 1990's [10, 11]. A review of the following surge in optical nanoscopy techniques can be found in [5, 12, 13] where we differentiated the so-called super-resolution techniques from those that use nanodetection. Nanodetection techniques allow the study of nanostructures without necessarily resolving them. They include many techniques classed according to the methods used, such as contrast, phase and deconvolution.

Super-resolution techniques achieve real improved lateral resolution [5], such as I²M (combination of FM, interference illumination microscopy and ¹M, incoherent interference illumination microscopy) [14], the use of 4π illumination [15], SL (scattering lens) microscopy [16], FSL (far field superlens), the Pendry hyperlens [17] and the use of microspheres such as in SMON (submerged microsphere optical nanoscopy) [18, 19]. All these techniques are limited to 2D imaging.

Those that have been developed specifically for 3D measurement include SIM (structured illumination microscopy) [20] and TDM (tomographic diffraction microscopy) the tomographic mode of digital holographic microscopy (DHM) [21, 22]. In the latter technique, the improvement in lateral resolution is achieved using a synthetic numerical aperture by varying the illumination angle. In the reflection mode, 130 nm patterns on a Blu-ray® disc have been profiled in 3D at a wavelength of 475 nm and with an axial sensitivity of 5 nm. This latter technique is particularly interesting since it uses the phase of the light to give the extremely high axial resolution.

Other interference based microscopy techniques are the well-known phase shifting microscopy (PSM) and coherence scanning interferometry (CSI, or white light scanning interferometry, WLSI) that are now mature non-contact techniques for measuring surface roughness and microscopic 3D surface structure. They provide quantitative information over large areas with sub-nanometer axial resolution [2] using high sensitivity phase measurement of the wavefront by comparing it with that from light reflected from a reference mirror in a dedicated interference objective. The technique has been very successful in the characterization of very rough surfaces such as biomaterials [23] and colloids [24]. But the lateral resolution is limited by diffraction, as mentioned, to a value of about 200 nm [3]. Recently, we have succeeded in combining the 2D super-resolution qualities of the microsphere technique with the high axial resolution PSM technique of interference microscopy, using a Linnik configuration. At the same time the technique has also been used with a Mirau objective [25], resulting in a 2 to 4 fold improvement in lateral resolution compared with classical imaging, and with the technique of white light fringe envelope detection using a Linnik configuration [26]. 3D super-resolution using microspheres has also been achieved in digital holography [27]. In practice, achieving successful results with this technique is challenging, due to the presence of the microsphere in the interferometer arm. The principle of operation is also little understood.

In the present work, we present our own first nano-3D results using the Linnik configuration in combination with glass microspheres developed independently during the preparation of the aforementioned papers. We demonstrate that the photonic nanojet produced by the glass microsphere plays an important role in the improvement in lateral resolution. High resolution 3D imaging of square profile grating structures is achieved by imaging with white-light illumination through 24 µm diameter glass microspheres placed on top of the sample in front of the interference objective. Results are shown of 3D measurements in air of a 0.6 µm period grating that is not visible with the same objective without the microsphere and of a 1.2 µm period grating whose 3D measurements have been improved through the microsphere. Some first results of simulations of the interaction between the photonic jet and the grating lead to some basic understanding of the image formation.
2. 3D microscope system

The system used is a modified Leitz-Linnik interference microscope [28] using identical x50 (NA = 0.85) objectives and a PIFOC piezoelectric nanopositioner controlled in a closed loop with a capacitive position sensor for the Z-scanning of the fringes over the depth of the sample (figure 1). Images are acquired with a monochrome CMOS camera (PhotonFocus MV1-D2048-96-G2) with a Giga Ethernet connection. The measurement system is controlled by a PC equipped with an Intel® Xeon® CPU processor (2.40 GHz, 8 Go RAM) under a Windows 7 (64 bits) operating system. The control and analysis software was developed in-house under LabVIEW (version 2014, 64 bits, from National Instruments) combined with the IMAQ Vision module.

![Figure 1. Schematic layout of the Linnik interference system used for nano-3D measurements.](image1)

The microspheres are carefully placed on the sample (figure 2(a)) so that they are positioned between the objective and the area of the sample to be observed (figure 2(b)). The Linnik interferometer is adjusted first to produce high contrast fringes directly on the surface of the sample and then through the microsphere on the surface by modifying the path length difference between the two arms to match the coherence planes.

![Figure 2. The nano-3D configuration (a) positioning of microsphere on sample surface in front of objective in the Zeiss classical optical microscope and (b) schematic layout of the microsphere placed on the sample, under the objective.](image2)
The algorithm used to make the phase measurement was a modified version of the 5 phase step algorithm \((\pi/2)\), together with image averaging. The central region of interest was isolated and a phase discontinuity algorithm was used to unwrap the phase, followed by removal of the spherical form and light (3x3 pixels) median/low pass filtering to reduce the camera noise.

The surface shape was also measured using a Park XE70 AFM microscope in a vibration isolation chamber working in the non-contact mode. The tip used was a noncontact high frequency point probe with a tip radius of 2 nm and a width of 10 nm at a distance of 100 nm from the tip. The maximum field size is 50x50 µm for 256x256 pixels, with a lateral resolution \(R_{lat} = 0.012 \mu m \) to 0.195 \(\mu m\) depending on the field size.

3. 3D resolution standard and fringe observation

For the demonstration of 2D super-resolution below 0.5 \(\mu m\), several samples have been used in the literature: the periodic lines of a Blu-Ray DVD disk (spacing of 100 nm), the pores in an anodic aluminum oxide (AAO) sample (average diameter of 50 nm), individual adenoviruses (approximately 75 nm) [18, 19] and CD-ROM surface patterns [21].

![Figure 3](image)

**Figure 3.** Reference gratings observed with Zeiss optical microscope with an objective of x50 (NA = 0.55) directly and through 24 \(\mu m\) diameter microspheres, changing focus from (i) to (iii) with a zoom in (iv) from the area of (iii) for (a) 1.2 \(\mu m\) pitch and (b) 0.6 \(\mu m\) pitch.

The choice of the sample to illustrate 3D super-resolution is not simple. 3D structures that have been used in the literature are the periodic line structures of a stripped Blu-Ray DVD disk (spacing of 100 nm) and a VLSI STR10-1000P silicon test pattern, composed of 5 \(\mu m\) wide, 100 nm high square structures with a pitch spacing of 5 \(\mu m\) [22].

In the present work we used a set of reference gratings (prototype RS-N gratings from SiMETRICS GmbH) consisting of several sub-gratings with different pitch values, dry etched in a 10 mm x 10 mm Si wafer up to a depth of 190 nm. The gratings measured had pitch values of 1.2 \(\mu m\) and 0.6 \(\mu m\). The glass microspheres have a diameter of 24 \(\mu m\) (from Cospheric, size dispersion of 5\%, \(n = 1.5\)). The microspheres placed on the 1.2 \(\mu m\) and 0.6 \(\mu m\) pitch gratings can be observed in figure 3, moving from the focus on the grating in figure 3(i) through to focus on the virtual image plane through the microsphere in figure 3(iii). The images in figure 3(iv) show a zoom from figure 3(iii) of the super-resolved grating structures.
The results in figure 4 show a 24 µm diameter sphere on the 0.6 µm pitch grating using the Linnik setup in air with white light. A 2D super-resolved image of the grating can be observed through the sphere in figure 4(a) and the fringes superimposed on the grating in figure 4(b). The latter image shows perturbations in the fringes due to the depth of the grating, showing that there is an influence of the phase of the light from the grating surface through the microsphere.

![Image](a) ![Image](b)

**Figure 4.** Imaging through the 24 µm diameter microsphere in white light (a) directly, showing the 2D super-resolved image of a 0.6 µm pitch grating and (b) fringes superimposed on the image of the grating through the microsphere.

4. 3D measurements of Si gratings

The following results show measurements made on two of the different gratings with the Leitz-Linnik microscope with and without the microspheres. Measurements of the gratings are also made by AFM. A summary of the depth comparisons are given in Table 1 at the end of the section. Since the periods of the gratings are known, the magnification obtained through the microspheres is determined by the ratio between the period measured through the microspheres and the theoretical period value.

4.1. Measurements on 1.2 µm pitch grating

The measurements made on the 1.2 µm pitch grating under the Leitz-Linnik are possible directly without the sphere (figure 5(d),(e),(f)) but gives only a depth of 30 nm, compared with the nominal depth of 192 nm. Through the microsphere of 24 µm diameter, the grating is magnified by a factor of 4.5 and is much squarer, showing a depth of 150 nm (figure 5(a),(b),(c)), which is closer to that measured by AFM. Figure 6(b) shows the profile measured by AFM in the non-contact mode and reveals a pitch of 1.21 µm and a total depth of around 151 nm. It can be noticed that the depth profile measured is not square but that the sidewalls of the well appear inclined, together with a V shaped structure in the bottom of the well. The reasons why these details are not observed with the Leitz-Linnik could be due to a lack of lateral resolution in the direct measurements (figure 5(d),(e),(f)) or a difference between the positions observed in AFM and Leitz-Linnik through the microsphere (figure 5(a),(b),(c)).
Figure 5. Comparison of measurements of the 1.2 μm pitch grating through the 24 μm diameter microsphere in white light (a) 3D view (b) grey level image of surface heights (c) line profile: average height = 150 nm and directly without sphere with the x50 Leitz-Linnik microscope (d) 3D (e) grey level surface heights and (f) line profile: average height = 30 nm.
Figure 6. AFM measurements of the 1.2 µm pitch Si grating (a) 3D image of 10 µm x 1.5 µm area and (b) false colour image of surface heights and (c) line profile from (b); height between green arrows = 153 nm.

4.2. Measurements on 0.6 µm pitch grating

The measurements made on the 0.6 µm pitch grating under the Leitz-Linnik are not possible directly, being unresolvable, with $R_c = 0.55$ µm (Rayleigh criterion). Through the microsphere of 24 µm diameter, the grating is visible with a magnification of x3.65 and measurable (fringes resolved), giving a depth of 46 nm (figure 7). These results demonstrate sub-diffraction limited 3D imaging.

Figure 7. Comparison of measurements of the 0.6 µm pitch grating through the 24 µm diameter microsphere in white light (a) 3D view, (b) grey level image of surface heights and (c) line profile: average height = 46 nm.

The results of the AFM measurement are shown in figure 8(c), revealing a pitch of 0.633 µm and a total depth of around 58 nm.
Figure 8. AFM measurements of the 0.6 μm pitch Si grating (a) 3D image of 2.3 μm x 0.34 μm area, (b) false colour image of surface heights and (c) line profile from (b); height between green arrows = 58 nm.

The results of the depth measurements with the different techniques are given in Table 1 for the two grating pitches.

Table 1. Comparison of depth measurements of different gratings of RS-N standard using different techniques. Legend: (xx) cannot be measured.

| Grating pitch (μm) | Nominal depth (nm) | Depth AFM (nm) | Leitz-Linnik WL (λ = 760 nm) | Through 24 μm microsphere |
|--------------------|--------------------|----------------|-----------------------------|---------------------------|
|                    |                    |                | Depth direct (nm) | Magnification | Depth (nm) |
| 1.2                | 192                | 153            | 30                        | 4.5                      | 150        |
| 0.6                | 160                | 58             | xx                        | 3.65                     | 46         |

5. Results of simulations

To better understand the phenomena involved, 2D FEM simulations were performed for a glass microsphere (n = 1.5) in air illuminated with a TE plane wavefront having a wavelength in free space of 500 nm coming from the top in figure 11. The boundary conditions for the right, left and bottom of the figure are Perfectly Matched Layer (PML) absorbing boundaries. A photonic nanojet is a propagated beam concentrated beyond the diffraction limit in the near field of a dielectric microsphere [29]. The formation of the photonic nanojet 3.5 μm below the microsphere can be clearly observed in figure 11(a), at which point (maximum intensity) the transverse Full Width at Half Maximum is around one wavelength. By analogy with a classical lens this could be considered as being the image focus point. When the sphere is on the grating surface, since the latter (the object) is between the lens and the image focus, the sphere may therefore create a virtual image with a magnification of larger than one. This is in agreement with the experimental observations but with an image position and magnification different to the ones predicted by classical optics, which can be easily understood because of the object scale compared to the wavelength.

The simulation for the case of the microsphere placed on top of the 1.2 μm pitch grating is shown in figure 11(b) with a close up of the zone of interest in figure 11(c). The parameters used for Si were n = 3.42, σ = 10^{12} Sm^{-1}, with a reflected ratio (power air/silicon) of 30%.
Figure 11. Determination of the imaging properties through a 24 µm glass microsphere illuminated with a TE plane wave at $\lambda = 500$ nm (from the top) using 2D FEM simulation in air (a) formation of the photonic nanojet below the microsphere $|E_z|$, (b) intensity of the wavefront emerging from the microsphere after interaction with the 1.2 µm pitch Si grating (real part of $E_z$) and (c) zoom from (b).

The results of these simulations show the interaction of the photonic nanojet with the silicon grating below the microsphere (figure 11(b) and (c)). Between 4 and 5 periods of the 1.2 µm grating are illuminated in accordance with the number of periods observed experimentally. The exact explanation of the super-resolution effect in 2D imaging is as yet unclear. Coherence must probably be taken into account, as must be the high spatial frequencies of the evanescent waves located in the near field [19, 25]. Independently, the simulations of figure 11(b) and (c) show that because of the contact between the sphere and the object, the light collected appears to correspond to a numerical aperture near to 1.

6. Conclusions
The aim of this work was to make an initial exploration of the 3D super-resolving properties of glass microspheres placed between the objective and sample of a Linnik interferometer. The first results using 24 µm diameter glass microspheres positioned on square profile gratings of different pitches in Si in a white-light Leitz-Linnik interferometer show that lateral super-resolution can be obtained and implemented with phase shifting to measure the 3D shape of surfaces. The smallest grating having a pitch of 0.6 µm could be measured through the microspheres whereas it could not be measured directly with the classical system, demonstrating sub-diffraction limited imaging.

Some initial simulations show the highly focused photonic nanojet in the near field of the microsphere. We have demonstrated how this can explain why an imaging process takes place with a magnification of larger than one. The perturbation of the electromagnetic field near to the contact between the microsphere and the grating also shows a numerical aperture of around 1 in air.
Both of the 3D measurement techniques displayed certain artefacts. For the super-resolved 3D images through the microspheres, there was notably regions of blur, which is possibly due to non-optimized illumination conditions or non-uniformity of the microspheres. For the AFM measurements, there are possible artefacts in the measurements of the grooves due to the interaction between the tip shape and grating edges. The results nonetheless confirm that nano-3D super-resolved imaging is possible in a far field optical imaging system using dielectric microspheres placed between the objective and sample. This new technique opens new possibilities for high resolution characterization in nanomaterials and the biological sciences.

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8. References
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