Scattering Optics resolve Nanostructure

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ABSTRACT

Scattering of light is considered a nuisance in microscopy. It limits the penetration depth and strongly deteriorates the achievable resolution. However, by gaining active spatial control over the optical wave front it is possible to manipulate the propagation of scattered light far in the multiple scattering regime. These wave front shaping techniques have given rise to new high-resolution microscopy methods based on strong light scattering. This is based on the realization that scattering by stationary particles performs a linear transformation on the incident light modes. By inverting this linear transformation, one can focus light through an opaque material and even inside it. An extremely high resolution focus can be obtained using scatterers embedded in a high-index medium, where the diffraction limit for focusing is reduced by a factor \( n \). We have constructed a scattering lens made of the high-index material gallium phosphide (GaP) which is transparent over most of the visible spectrum and has the highest index of all nonabsorbing materials in the visible range. This yields a focal spot resolution of less than 100 nm, and it seems theoretically possible to create a focus of order 70 nm. The system resolution of a microscope based on this lens could be substantially higher.

**Keywords:** Imaging, Microscopy, Nanofabrication, Semiconductors, Light Scattering

1. INTRODUCTION

It is well known that optical elements in microscopes and cameras need to be kept impeccably clean as any scattering from impurities will result in a deteriorated image. Likewise, lens and mirror surfaces are manufactured with high precision to avoid aberrations that cause a loss of resolution. Nonetheless, unavoidable fabrication imperfections cause disorder in optical components that affects the image quality. On top of that, disordered light scattering by the environment strongly limits imaging inside turbid materials, such as biological tissue.\textsuperscript{1,2} To counteract the disorder, gated imaging methods, such as optical coherence tomography, quantum coherence tomography,\textsuperscript{3} time gated imaging,\textsuperscript{4} and polarization gated imaging\textsuperscript{5} reject scattered light to improve the imaging depth. As the fraction of ballistic light exponentially decreases with increasing depth, the use of these techniques is limited to a few mean free paths. On the other hand, methods such as diffuse optical tomography\textsuperscript{6} that use the multiple scattered light to form an image, can work at great depth but have poor resolution.

The insight that scattering does not have to be detrimental to the resolution came from Freund\textsuperscript{7} who considered experiments with light waves in the mesoscopic regime. First experimental demonstrations were made in the fields of acoustics and microwaves where these classical waves shown to be a convenient testing ground of mesoscopic wave physics. By taking advantage of the time reversal invariance, the group of Fink demonstrated that a pulse can be focused back onto its source by reradiating a time reversed version of the scattered wave front.\textsuperscript{8,9} This powerful concept was used to focus waves through disordered barriers,\textsuperscript{10-12} increase the information density in communication,\textsuperscript{13-16} and even address receivers below the diffraction limit\textsuperscript{16,17} with the use of disordered scattering. These methods are based on digital recording and playback of the acoustic or RF field, which cannot be directly applied in the optical regime.

In 2007, our group demonstrated the first focusing of multiple scattered light, which was achieved by controlling the incident wavefront.\textsuperscript{18} Subsequently, wavefront control methods were employed to focus light deep inside...
Figure 1. Comparison of light focusing with a conventional lens and a scattering lens. (a) A plane light wave sent through a normal lens forms a focus. The focal size is determined by the range of angles in the converging beam as and by the refractive index of the medium that the light is propagating in. The microscope image shows a collection of gold spheres as imaged with a commercial high quality oil immersion microscope objective. Inset on left is a photo of a conventional glass lens. (b) A shaped wave sent through a scattering layer on top of a high refractive index material. The wave front is carefully shaped so that, after traveling through the layer, it forms a perfectly spherical, converging wave front. The large range of angles contributing to the converging beam, combined with the high refractive index, give rise to a 97-nm resolution focal spot. The microscope image shows the same collection of gold spheres as in (a) imaged with the scattering lens. Inset on left is a photo of the lens with the scattering layer on top.

strongly scattering materials,\textsuperscript{19} to study basic transport physics,\textsuperscript{20} to create optical traps behind scattering materials,\textsuperscript{21} to measure the optical transmission matrix\textsuperscript{22} and to transmit images through turbid layers.\textsuperscript{23} Wavefront control is closely related to phase conjugation\textsuperscript{24} in turbid materials,\textsuperscript{25} and a digital optical phase conjugation method that is based on the principles of wavefront control\textsuperscript{26} has great promise for use in biomedical context.\textsuperscript{26} The combination of digital phase conjugation with nonlinear optical nanoprobes offers a realistic prospect for in-tissue imaging.\textsuperscript{27}

Remarkably, the size of the focus that is formed by wavefront control of scattered light is independent of the numerical aperture or even the quality of the optics through which the incident light passes. Only the numerical aperture for propagation from the scatterers to the target determines the resolution, which can therefore be much higher than that of the wavefront shaping optics.\textsuperscript{14,28} Based on this principle, we recently demonstrated a scattering lens which has a freely scannable 97-nm focus of visible light.\textsuperscript{29} In this lens, the principle of resolution enhancement by scattering is used in conjunction with the high-index solid immersion medium gallium phosphide (GaP), a combination termed High-Index Resolution Enhancement by Scattering (HIRES). In the present paper, we present details on the implementation of the HIRES scattering lens, its relation to other high-resolution optical methods and the prospects for applying scattering optics in science and technology.

2. HIGH INDEX RESOLUTION ENHANCEMENT BY SCATTERING

While near-field techniques and the related superoscillating waves\textsuperscript{30} are not subject by any fundamental limit in resolution,\textsuperscript{31} far-field approaches can not resolve arbitrary small objects. In fact the smallest possible focus is given by the Abbe diffraction limit \(d \geq (0.47\lambda)/\text{NA}\), where \(\lambda\) is the wavelength and NA the numerical aperture. Note that we use Sparrow’s resolution criterion in calculating the spot resolution. Reducing the wavelength increases the resolution but there are often practical limits to this approach making the increase of the numerical aperture the path of choice to improve the resolving power.

Oil immersion microscopes use an index-matched liquid to increase the medium refractive index and thus boost NA, and deliver resolution down to 160 nm. Using a solid medium between the optics and the object to be imaged allows for higher refractive indices and thus for a better resolution,\textsuperscript{32,33} but this approach requires a very careful alignment of the solid material and is very sensitive to any manufacturing imperfections. Moreover, most
crystalline solids have intrinsic birefringence. In case of cubic crystals, complicated intrinsic birefringence\textsuperscript{34,35} distorts the wavefronts in a manner that is hard to predict or compensate, so that the GaP immersion lenses that have been realized\textsuperscript{36–40} have so far not reached 100-nm resolution.

The HIRES lens circumvents most issues related to lens fabrication and characterization. A slab of a dielectric material is chemically etched on one side to create a scattering layer (acting as a lens) and the object to be measured is placed on the other side. The disordered layer scramble the incident light coupling the light to all the propagating modes inside the slab. In order to take advantage of it the material used must have a high refractive index, have negligible absorption and must work at the shortest possible wavelength.

In Fig. 2 we show the refractive index at the lowest transparency wavelength (defined as the wavelength where the imaginary part of the refractive index reaches $10^{-3}$) for a number of common materials. The best possible resolution of a HIRES lens (or in fact any immersion lens) is given by the diffraction limit at this wavelength. Lines of constant diffraction limit are shown. The highest resolution of the indicated materials can be obtained with aluminum arsenide (AlAs), however, this toxic material is unstable in contact with water. Gallium phosphide offers a resolution that is only slightly lower with much better chemical stability. At a wavelength of 561 nm GaP has a refractive index of $n = 3.41$ and is completely transparent, giving a GaP HIRES lens the potential to reach a 72 nm resolution.

3. CONSTRUCTION AND OPERATION

A HIRES-lens consists of a homogeneous slab of high-index material on top of a strongly disordered scattering layer. The disordered layer breaks the translational invariance of the interface, which enables incident light to be coupled to all propagating angles inside the high-refractive-index material. Yet multiple scattering also scrambles the wavefront creating a speckle-like pattern on the object plane that itself cannot be used for imaging. Therefore we manipulate the incident wavefront in order to force constructive interference of the scattered light at a position in the object plane of our HIRES-lens. The wavefront is controlled using a feedback based method.\textsuperscript{18} As a result, a perfectly spherical wave emerges from the porous layer and converges towards the object plane to form a sharp...
A $\lambda_0 = 561$ nm laser beam is expanded and illuminates a phase-only spatial light modulator. The modulated reflected beam is first imaged onto a two-axis steering mirror and then onto the porous surface of the GaP HIRES-lens. A variable aperture controls the extent of the illuminated area and a light stop places the setup in a dark field configuration by blocking the center of the light beam. We image the object plane onto a CCD camera using an oil immersion microscope objective.

An overview of our setup is shown in Fig. 3. We use a continuous wave laser with a wavelength of $\lambda_0 = 561$ nm just below the GaP bandgap of 2.24 eV (550 nm) where the refractive index is maximal and the absorption is still negligible. We spatially partition the wavefront into square segments of which we independently control the phase using a spatial light modulator (SLM). The SLM is first imaged onto a two-axis fast steering mirror and then onto the porous surface of the HIRES-lens. With a variable aperture we set the radius $R_{\text{max}}$ of the illuminated surface area between 0 $\mu$m and 400 $\mu$m. The visibility of the gold nanoparticles is maximized by blocking the central part of the illumination ($R < 196 \mu$m), placing the system in a dark field configuration. At the back of the HIRES-lens a high-quality oil immersion microscope objective (NA = 1.49) images the object plane onto a CCD camera. This objective is used to efficiently collect all the light scattered from the object plane and to obtain a reference image.

4. ANTI-INTERNAL-REFLECTION COATING

Due to the high refractive index contrast at the GaP/air interface, a large fraction of the light is internally reflected. Such detrimental reflections also occur in solid immersion lenses, in the HIRES lens their effect is to severely decrease the optical memory range. We developed an anti internal-reflection (AIR) coating to suppress these interfering reflections. The coating consists of an approximately 200 nm thick layer of amorphous silicon (Si) deposited on top of the HIRES-lens by electron-gun evaporation. The amorphous Si, which has a tabulated refractive index of $n = 4.8$ and an absorption length of $\ell_{\text{abs}} = 0.94 \mu$m, is close to index matched with the GaP and strongly absorbs the light that would otherwise be internally reflected.

To test how well our AIR coating functions, we monitor the correlation of the scattered field while we rotate the beam incident on the porous layer. For this experiment we removed the light stop from the setup, enabling a direct recording of a speckle pattern on the CCD camera. In Fig. 4 we show the results for an uncoated and a coated GaP HIRES-lens. For the uncoated case, a large correlation loss of 76% is seen for a small angular change of the incident field. After the initial decline, the correlation continues to decrease according to a functional form predicted by the optical memory effect. When we measure the correlation loss in a coated HIRES-lens, we see that decrease is almost completely governed by the memory effect. The large loss seen in the uncoated
Influence of a Si anti internal-reflection coating on the correlation range of scattered light inside a GaP HIRES-lens when we change the angle $\theta$ under which the beam incidents. The correlation range determines the Field of View of the lens and should therefore be as large as possible. In an uncoated GaP HIRES lens internal reflections significantly reduce the correlations. By coating the lens with a thin layer of amorphous Si these reflections are suppressed resulting in a much larger correlation range. The green solid line represents theoretical loss of correlation due to the optical memory effect.

Figure 5. Schematic representation of light steering in a HIRES-lens. The vertical position on the porous layer is defined as $x$ and the vertical position in the object plane of the lens is defined as $u$. The homogeneous part of the HIRES-lens has a thickness $L$. The focus, which is initially created on position $u = 0$, is scanned towards $u = u_0$.

The lens is now reduced to a marginal 4%. The lines through the data points represents the theoretical expected decorrelation. We used the porous layer thickness as a free parameter. These results show that the AIR coating is capable of properly eliminating the unwanted internal reflections inside the GaP HIRES-lens.

By placing a small droplet of photoresist (Olin 908/35) on top of GaP before we deposit the Si AIR coating, we were able to lift off an area of the coating afterwards. This process creates a window in the coating in which we place objects onto the HIRES-lens.

5. LIGHT STEERING

The high-resolution focus can be scanned across the surface of the lens by exploiting the angular correlations in the scattered light known as the optical memory effect. To steer the focus to a new position, we add an additional phase to the incident wave front. A schematic representation of the HIRES-lens is given in Fig. 5. We will derive the required phase in one dimension which then is easily generalized for the two-dimensional case.

When the initial focus is created, we force light coming from every illuminated position $x$ on the porous layer to arrive with the same phase at position $u = 0$. If we now want to move the focus to $u = u_0$, we need to compensate the phase for the path length difference between the original and the new position. The required
Figure 6. (A) The calculated intensity profile of the focus. (B) A collection of gold nanoparticles imaged with the GaP HIRES-lens. The image contains the scattering intensity as a function of the focus position. Due to the profile of the focus, side lobes are visible around the nanoparticles. (C) Spectral contributions of the transversal k-vectors $k_{\text{trans}}$ of the calculated focus intensity (blue line) and the acquired intensity image (red bars). The scale bars represent $300 \text{ nm}$.

The phase $\phi$ depends on the position on the porous layer $x$ and is given by

$$\phi(x)_{u_0} = \left[ \sqrt{(x - u_0)^2 + L^2} - \sqrt{x^2 + L^2} \right] \frac{k}{2}$$

$$= -\frac{xu_0}{\sqrt{x^2 + L^2}} k + \mathcal{O}(u_0^2),$$

(1)

where $L$ is the thickness of the homogeneous layer and $k$ is the wave number of the light. The final result is obtained by a Taylor expansion around the original focus position $u = 0$. Generalizing Eq. 1 to two dimensions results in the required phase offset at position $(x, y)$ to move the focus in the object plane towards position $(u_0, v_0)$

$$\phi(x, y)_{u_0, v_0} \approx -\left[ \frac{xu_0}{\sqrt{x^2 + L^2}} + \frac{yu_0}{\sqrt{y^2 + L^2}} \right] k.$$  

(2)

When the illuminated surface area is much smaller than the thickness of the HIRES-lens, the required phase reduces to a linear phase gradient. For our experiments however, the illuminated surface area is of the same order as the thickness $L$ making it necessary to use Eq. 2.

6. IMAGE ACQUISITION AND PROCESSING

As a test sample we have placed gold nanoparticles on top of the window in the coating. These particles were deposited by first placing $5 \mu L$ of solution, which contained $50$-nm diameter gold nanoparticles (BBInternational GC 50). After the solvent evaporated, a random collection of gold nanoparticles with a density of $1$ sphere/$\mu m^2$ was left on the object plane of the HIRES-lens.

The shape of the focus generated in our HIRES-lens is related directly to the illuminated area of the porous layer. To maximize the contrast of the nanoparticles, we blocked the central part of the illumination beam. As a result the side lobes of our focus become more pronounced as is shown in Fig. 6A. By scanning this focus...
over the gold nanoparticles in the object plane of the HIRES-lens, we acquire an image that is the convolution of the focus with the structure (Fig. 6B). In this raw scan result we see the influence of the side lobes as rings around the nanoparticles. To remove these rings, we divide the spectral spatial components of the image with the calculated spatial spectrum of the focus. The spectral contributions of the transversal k-vectors $k_{\text{trans}}$ of the focus intensity and the image intensity are shown in Fig. 6C. The maximal contributing transversal k-vector $k_{\text{max}} \approx 8/2\pi$ suggests an even higher possible resolution than the 97-nm value we obtained so far.

7. PROSPECTS

While a HIRES lens can provide a significantly higher resolution than conventional optics, its resolution is finally limited by the availability of relatively few transparent high-index materials. Recently, many viable approaches to obtaining high resolution imaging with visible light have been proposed and demonstrated. High resolution can be obtained for dye-doped structures exploiting the photophysics of extrinsic fluorophores. Their resolution strongly depends on the shape of the optical focus, therefore a combination with a GaP HIRES lens could possibly yield an even higher resolution if the object to be imaged is close to the surface. Pulsed light is a requirement for some of these methods, therefore the recent expansion of wavefront control methods to femtosecond pulsed light is an important step.

Other imaging methods reach high resolution by reconstructing the evanescent waves that decay exponentially with distance from the object. Near field methods reach a very high resolution and recording density by bringing nano-sized scanning probes or even antennas in the close proximity of the sample. It will be interesting to combine HIRES lenses with near-field probes which could characterize the field in the vicinity of the focus. Possibly, a near-field probe could be used to calibrate a HIRES lens or vice versa.

Metamaterials, which are meticulously nanostructured artificial composites, can be engineered to access the evanescent waves and image sub-wavelength structures as demonstrated with superlenses and hyperlenses for ultraviolet light. These materials physically decrease the focus size, which will lead to improvement of linear and non-linear imaging techniques. By constructing a metamaterial with a high index in the visible range, the resolution of a HIRES lens could be pushed further. In the especially important visible range of the spectrum, plasmonic metamaterials can be used to produce nano-sized isolated hot spots which are very promising tools for high-resolution imaging. So far the inability to scan these hotspots limits their usefulness for microscopy, a problem that may in future be resolved using recently developed flexible methods to displace a plasmonic focus. Remarkably, spheres of relatively low-index materials have been used to obtain images at surprisingly high resolution, resolving holes in a metal film with a 100-nm center-to-center distance, albeit with a small field of view. Nano-patterned substrates in the near-field of the object may improve the imaging resolution similarly. In all these approaches, it will be beneficial to already inject light at very high resolution using the HIRES lens.

Higher resolution can also be obtained if knowledge on the shape of the object is available. In these cases the image can be numerically reconstructed starting from a low resolution measurement and imposing some constraints. These methods effectively extrapolate high spatial frequency information from the measured low spatial frequencies. Obviously, their resolution depends on the base resolution with which the underlying image information is acquired, and these methods could be improved in combination with a HIRES lens.

Nano-patterning substrates in the near-field of the object is another method for increasing the imaging resolution, and resonant spheres of relatively low-index materials have been used to obtain images at surprisingly high resolution, resolving holes in a metal film with a 100-nm center-to-center distance, albeit with a small field of view. These approaches strongly depend on a multiple scattering interaction on nanostructures near the object, and therefore may not be easily generalized. They suggest that even higher resolution may be obtained by randomly or periodically structuring the object plane of the HIRES lens.

Finally, since a HIRES lens is a linear optical element it preserves the phase of the light field and can therefore be used in phase sensitive microscopy methods such as $4\pi$ microscopy or optical diffraction tomography. In the latter case, the resolution could possibly be improved to 50 nm or better. Finally, a wide-field high-speed version of the HIRES lens could be based on a very recently demonstrated fast high-resolution scattering lens method.
8. CONCLUSIONS

We have shown the realization and analyzed the performance of a HIRES lens, a new type of lens based on scattering and a high-index medium. It presently offers the highest-resolution scannable focus of visible light, just below 100 nm, which makes it possible to study nanostructures with visible light. The system resolution of a modern microscopy method employing a HIRES lens could be even much below 100 nm. Combination of scattering-based lenses with existing high-resolution microscopy methods is likely to significantly extend the possibilities of high-resolution imaging. This will lead to new applications of high resolution microscopy in regions where the complexity and fragility of current systems was forbidding so far.

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