Scattering Lens Resolves Sub-100 nm Structures with Visible Light

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The smallest structures that conventional lenses are able to optically resolve are of the order of 200 nm. We introduce a new type of lens that exploits multiple scattering of light to generate a scanning nanosized optical focus. With an experimental realization of this lens in gallium phosphide we imaged gold nanoparticles at 97 nm optical resolution. Our work is the first lens that provides a resolution better than 100 nm at visible wavelengths.

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Many essential structures in nanoscience and nanotechnology, such as cellular organelles, nanoelectronic circuits, and photonic structures, have spatial features in the order of 100 nm. The optical resolution of conventional lenses is limited to approximately 200 nm by their numerical aperture and therefore they cannot resolve nanostructure. With fluorescence based imaging methods it is possible to reconstruct an image of objects that are a substantial factor smaller than the focus size by exploiting the photophysics of extrinsic fluorophores [1–5]. Their resolution strongly depends on the shape of the optical focus, which is determined by conventional lens systems. This dependence makes them vulnerable to focal distortion by scattering. Moreover, it is not always feasible to dope the object under study. Other imaging methods reach high resolution by reconstructing the evanescent waves that decay exponentially with distance from the object. Near field microscopes [6] reach a very high resolution by bringing nanosized scanning probes or even antennas [7] in close proximity of the object to detect the evanescent field. A drawback is that the scanning probe may interact with some samples and perturb their structure. Metamaterials, which are meticulously nanostructured artificial composites, can be engineered to access the evanescent waves and image subwavelength structures [8] as demonstrated with superlenses [9] and hyperlenses [10] for ultraviolet light. These materials physically decrease the focus size, which will lead to improvement of linear and nonlinear imaging techniques. In the especially important visible range of the spectrum, plasmonic metamaterials can be used to produce nanosized isolated hot spots [11–13], but the limited control over their position makes their use for imaging a challenge. So far, a freely scannable nanosized focus of visible light has not been demonstrated.

In this Letter we introduce a new type of lens that generates a scanning sub-100 nm optical focus. We used this lens to image a collection of gold nanoparticles at 97 nm optical resolution. The lens exploits multiple scattering of light in a porous high-refractive-index material to increase the numerical aperture of the system, a principle we name high-index resolution enhancement by scattering (HIRES).

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 as shown in Fig. 1(a). Yet multiple scattering also scrambles the wave front creating a disordered intensity pattern resembling laser speckle on the object plane that itself cannot be used for imaging. Therefore we manipulate the incident wave front in order to force constructive interference of the scattered light at a position in the object plane of our HIRES lens. The wave front is controlled using a feedback based method [14] that is conceptually related to phase conjugation [15] and time

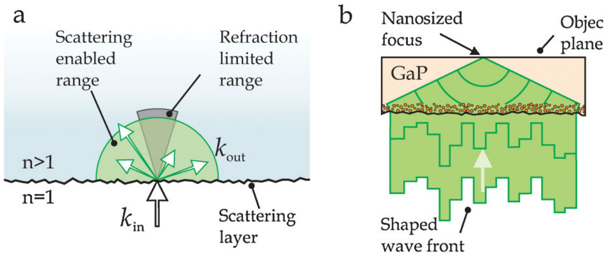

FIG. 1 (color). (a) Principle of light coupling to high transversal k vectors into a high-index material. Without scattering refraction would strongly limit the angular range to which light could be coupled. By exploiting strong scattering at the interface, incident light k_{in} is coupled to all outgoing angles k_{out} in the high-index material. (b) Schematic of a HIRES lens that uses light scattering to achieve a high optical resolution. This HIRES lens consists of a slab of gallium phosphide (GaP) on top of a strongly scattering porous layer. By controlling the incident wave front, a small focus is made in the object plane of the HIRES lens.

reversal [16]. As a result, a perfectly spherical wave emerges from the porous layer and converges towards the object plane to form a sharp optical focus [Fig. 1(b)]. Whereas in conventional optics (e.g., solid immersion lenses [17] or total internal reflection microscopes [18]) any inevitable surface roughness causes a distortion of the wave front and a concomitant loss of resolution, the inherent random nature makes a HIRES lens robust for these aberrations. Any wave front error is distributed randomly over all outgoing directions, reducing the contrast but not the resolution [19]. Therefore, a HIRES lens reaches the resolution of an ideal GaP immersion lens, which is much better than the resolution reached by even the best GaP immersion lenses [17] so far. In order to use the HIRES lens for high-resolution imaging, the focus is moved around in the object plane by steering the incident wave front, directly exploiting the angular correlations in the scattered light known as the optical memory effect [20–22]. By raster scanning the focus across an object we acquire an aberration-free high-resolution image. The robust scanning high-resolution focus makes the HIRES lens excellently suited for optical imaging of nanostructures.

To demonstrate an experimental implementation of our HIRES lens we fabricate it in gallium phosphide (GaP). GaP is transparent in a large part of the visible spectrum ($\lambda_0 > 550$ nm) and has a maximum refractive index of $n = 3.41$, higher than any other transparent material in this wavelength range [23]. Electrochemically etching GaP with sulfuric acid (H_2SO_4) creates macroporous networks resulting in one of the strongest scattering photonic structures ever observed [24]. Using this etching process we create a $d = 2.8$ μm thick porous layer on one side of a crystalline GaP wafer. This layer is thick enough to completely randomize the incident wave front and to suppress any unscattered background light.

The memory effect allows us to shift the scattered light in the object plane of the HIRES lens over a distance $r \approx 1.8L\lambda/(2\pi n^2 d)$ before the intensity correlation decreases to $1/e$ [20], where $L = 400$ μm is the thickness of the wafer. The loss of correlation only affects the intensity in the focus (not its shape) making it easy to correct for this effect without losing resolution. Because of the high-refractive-index contrast on the flat GaP-air interface, a large fraction of the light is internally reflected. The reflected light interferes with the light that comes directly from the porous layer. This interference causes a background signal that is 3 times larger than the focus intensity. To strongly suppress the internal reflections we have deposited an approximately 200 nm thick anti-internal-reflection coating of amorphous silicon on the surface. The amorphous silicon is nearly index matched with the GaP and strongly absorbs the light that would otherwise be internally reflected. As a result of this layer, the background signal is reduced to only 0.04 times the focus intensity [25]. The resulting field of view of our coated HIRES lens is measured to be $r = 1.7 \pm 0.1$ μm in radius, 85% of the theoretical limit determined by the optical memory effect. In the center of the surface we created a small window of about 10 μm in diameter by locally removing the anti-internal-reflection coating. We use this window to place objects onto our HIRES lens. As a test sample we have deposited a random configuration of gold nanoparticles with a specified diameter of 50 nm inside this window.

An overview of our setup is shown in Fig. 2. We use a continuous wave laser with a wavelength of $\lambda_0 = 561$ nm just below the GaP band gap of 2.24 eV (550 nm) where the refractive index is maximal and the absorption is still negligible [23]. We spatially partition the wave front 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 $R_{\max}$ of the illuminated surface area between 0 and 400 μm. The visibility of the gold nanoparticles is maximized by blocking the central part of the illumination ($R < 196$ μm), 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.

We first synthesize the wave front that, after being scattered, creates a focus in the object plane. We use light scattered from one of the gold nanoparticles in the object plane as a feedback signal to obtain a set of complex amplitudes that describe the propagation from different incident positions on the porous layer towards the nanoparticle [14], which can be interpreted as transmission matrix elements [26]. By reversing the phase of these

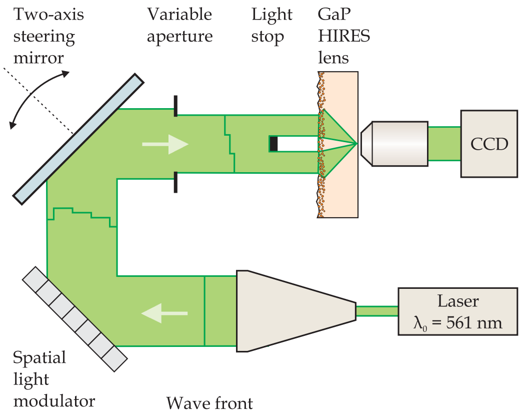

FIG. 2 (color). Overview of the setup. A $\lambda_0 = 561$ nm laser beam is expanded and illuminates a phase-only spatial light modulator. The modulated reflected beam is first imaged onto a two-axis steering mirror and then onto the porous surface of the GaP HIRES lens. A variable aperture controls 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.

complex amplitudes we force the light waves to interfere constructively at the exact location of the nanoparticle. The focus is moved around in the image plane by rotating every contributing k vector over a corresponding angle. We apply these rotations by adding a deterministic phase pattern to the incident wave front. In the paraxial limit, a simple tilt of the wave front would suffice to displace the focus [22,27]. For our high-resolution focus, which lies beyond this limit, an additional position dependent phase correction is required that we apply using the SLM. The total phase ϕ required at position (x, y) to move the focus towards position (u_0, v_0) in the object plane is given by [25]

$$\phi(x, y)_{u_0,v_0} = -\left[\frac{xu_0}{\sqrt{x^2 + L^2}} + \frac{yv_0}{\sqrt{y^2 + L^2}}\right]k + \mathcal{O}(u_0^2, v_0^2).$$

The addition of this correction is essential for a proper displacement of the focus.

In Fig. 3 we show the imaging capabilities of the GaP HIRES lens. First a reference image was acquired with the high-quality microscope behind the HIRES lens [Fig. 3(a)]. Because the gold nanoparticles are much smaller than the resolution limit of this conventional oil immersion microscope the image of the nanoparticles is blurred. Next we used our HIRES lens to construct a high-resolution image. By manipulating the wave front a focus was generated on the leftmost nanoparticle. We raster scanned the focus across the object plane while we recorded the amount of scattered light. In Fig. 3(b) the result of the scan is shown [25]. A cross section through the center of the left sphere [Fig. 3(c)] clearly shows the improvement in resolution we obtained with our HIRES lens, confirming our expectations that the resolution of this image is far better than that of the conventional high-quality detection optics.

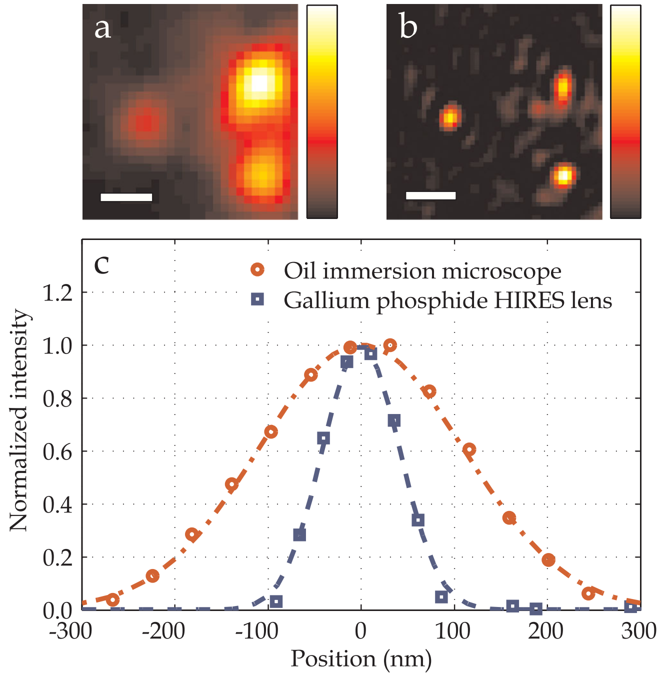

FIG. 3 (color). Experimental imaging demonstration with a GaP HIRES lens. (a) A reference image taken with conventional oil immersion microscope (NA = 1.49). The image shows a blurred collection of gold nanoparticles. The scale bar represents 300 nm. (b) A high-resolution image acquired with our GaP HIRES lens. The image was obtained by scanning a small focus over the objects while monitoring the amount of scattered light and deconvoluted with Eq. (1) [25]. (c) A vertical cross section through the center of the left sphere in (a) and (b) shows the increase in resolution. The dashed lines are Gaussian fits to the data points.

For a more quantitative study of the obtained resolution, we study the shape of the focus in the HIRES lens. The radial intensity distribution of the focus is directly calculated from a plane wave decomposition of the contributing waves,

$$I(r) = I_0\left[k_{\max}^2 \frac{J_1(k_{\max} r)}{k_{\max} r} - k_{\min}^2 \frac{J_1(k_{\min} r)}{k_{\min} r}\right]^2, \tag{1}$$

where J_1 is a Bessel function of the first kind. The minimum and maximum coupled transversal k vectors, $k_{\min}$ and $k_{\max}$, are directly related to the inner and outer radius, $R_{\min}$ and $R_{\max}$, of the illuminated area: $k_{\max} = nk_0(1 + L^2/R_{\max}^2)^{-(1/2)}$ (and similar for $k_{\min}$). To confirm this dependence, we imaged the objects for different values of the illumination radius $R_{\max}$. For each measurement the resolution is determined by modeling the resulting image of a single 50 nm gold nanoparticle with Eq. (1). Since it is hard to quantify the resolution from the width of a non-Gaussian focal shape we use Sparrow's criterion which defines the resolution as the minimal distance at which two separate objects are still discernible; see, e.g., [28]. In Fig. 4 the measured resolution versus $R_{\max}$ is shown. As a reference we also plotted the measured resolution of the oil immersion microscope. We see that the resolution improves as we increase the illuminated area. The measured

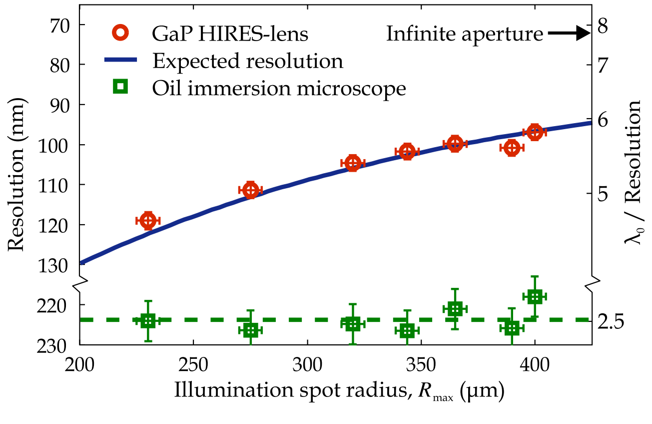

FIG. 4 (color online). Optical resolution of a GaP HIRES lens for different radii $R_{\max}$ of the illumination area. Red circles: measured resolutions of the HIRES lens. Solid blue line: expected theoretical resolution deduced from Eq. (1). Green squares: measured resolution of the oil immersion microscope. Dashed green line: mean measured resolution. Black arrow: expected resolution for an infinitely large illumination area.

resolutions are in excellent correspondence with the expected resolution obtained from the calculated intensity profile. The resolution of the HIRES lens is much better than the high-quality conventional oil immersion microscope. The highest resolution we measured is 97 ± 2 nm, which demonstrates imaging in the nanometer regime with visible light.

A GaP HIRES lens has the potential to reach even better optical resolutions up to 72 nm. It is then possible to resolve objects placed in each others near field at distances of $\lambda_0/2\pi$. To achieve these resolutions a wider area of the scattering porous layer has to be illuminated and as a result light has to be scattered at increasingly higher angles from the porous layer. Here advances could benefit from investigations in the field of thin film solar cells where high angle scattering is beneficial for optimal light harvesting [29].

Our results promise to improve resolution in a wide range of optical imaging techniques. The robustness of a HIRES lens against distortion and aberration, together with its ease to manufacture, makes it ideal for the imaging of fluorescent labeled biological samples or for the efficient coupling to metamaterials [9,10] and plasmonic nanostructures [11–13]. Recent developments in spatiotemporal control of waves in disordered materials [30–32] indicate the possibility for HIRES lenses to create ultrashort pulses in a nanosized focus. Since a HIRES lens is a linear and coherent optical element it is possible to combine it with interferometric methods for high precision field measurements [33,34] and to use it for resolution improvement of a large range of imaging techniques, such as confocal microscopy, STED [2], PALM [3], and STORM [4].

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