Unusual imaging properties of superresolution microspheres

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Abstract: We employ a self-assembly method to fabricate dielectric microsphere arrays that can be transferred to any desired positions. The arrays not only enable far-field, broad-band, high-speed, large-area, and wide-angle field of views but also achieve superresolution reaching $\lambda/6.4$. We also find that many proposed theories are insufficient to explain the imaging properties; including the achieved superresolution, effects of immersion, and unusual size-dependent magnification. The half-immersed microspheres certainly do not behave like any ordinary solid immersion lenses and new mechanisms must be incorporated to explain their unusual imaging properties.

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1. Introduction

According to the Rayleigh criterion of the limit of resolution, a conventional microscope cannot distinguish two objects closer than \( 0.61\lambda/NA \) (where \( \lambda \) is the wavelength of the incident light, \( NA \) is the numerical aperture of the objective). Thus for a typical microscope objective with \( NA \sim 1 \), the diffraction limit is \( \sim 0.61\lambda \). During the past decades, great advancements have been made to resolve features smaller than the diffraction limit. These superresolution techniques generally employ nonlinear optical effects or evanescent waves to overcome the diffraction limit [1]. Unfortunately, one always encounters drawbacks from high laser intensity irradiation, short lifetime of fluorescent dyes, or limited imaging speed for the nonlinear optical techniques based on switching or saturation of fluorescent emission. For the techniques enabled by evanescent waves, near-field scanning optical microscopy, superlens, or far-field hyperlens also have disadvantages on the scanning speed, sophisticated fabrication processes, or limited choices of samples [2]. To make widespread impact to other branches of sciences, it is highly desired to make the superresolution techniques easily accessible and low-cost.

In 2011, Z. B. Wang et al reported that dielectric microspheres are capable of reaching 50nm resolution using an ordinary white-light microscope [3]. Superresolution was found in imaging metallic structures as well as insulators using ordinary light sources [3–7], suggesting that neither nonlinear optical effects nor surface plasmons play any roles here. Later, the same group reported that an unprecedented \( \lambda/17 \) spatial resolution can be reached when combining it with a confocal microscope [7]. Surprisingly, many drawbacks of the established superresolution techniques so far seem to have been overcome by the microspheres with ease.

Yet it must be pointed out that not many of the observed properties are well understood or carefully characterized. For example, some of the previous superresolution claims were based on the smallest feature sizes rather than the deconvolution analyses, rendering over-estimation of the resolution capabilities [5,8–11]. Additionally, it remains a puzzle why some groups reported that half-immersed microspheres yield better resolution [8,12], while others claimed that sub-diffraction images can be obtained without employing any immersion media [5,9]. Moreover, unlike conventional lens, the magnification of the image was found to be diameter-dependent, deviating from the result of ray optics [9,13,14]. Furthermore, many imaging properties of barium titanate glass (BTG, whose refractive index \( n = 1.9-2.1 \)) microspheres appear to be characteristically different from those of others and the reasons for these remain unclear [6,11,13–15]. Finally, when imaging non-metallic samples, why the images of anodic aluminum oxides seem to yield better resolution (for example, the resolution is better than 25nm in [7]) than other structures [4,11]?

All of the unusual properties have stimulated researchers to look for a consistent and satisfactory explanation. At the beginning, numerical simulations or calculations based on
Mie theory suggested a subwavelength photonic nanojet forming at the focal point of a microsphere [9,16]. However, the width of the nanojet was numerically and experimentally found to be $\sim \lambda/2$ [17,18], much larger than the claimed resolution. Later, it was suggested that a resonant nanojet with a $\sim \lambda/3$ width could appear once the incident wave satisfies the resonant condition of whispering gallery modes of the microspheres [19–21]. As will be discussed later, the proposal is still insufficient to explain the achieved resolution and many other imaging properties.

From applications’ point of view, current superresolution techniques often require laser excitations and dedicated image processing. Thus many superresolution microscopes are not portable and their fields of view are limited to a few micrometers. In addition, the cost of superresolution microscopes usually exceeds US$1,000,000, inhibiting their widespread applications. On the other hand, the material cost of glass microspheres is less than US$1 per kilogram. The low-cost and easy-access are indeed two unmatched advantages of superresolution microspheres. To further widen their potential applications at large scales, it is desirable to develop a method that can enable quick assembling of microsphere arrays, and at the same time, keep the cost of the fabrication low.

In this paper, we apply rigorous deconvolution analyses and determine that the best spatial resolution of half-immersed microspheres can reach $\lambda/6.4$. We find that half-immersed microspheres always yield better resolution than other geometries. We further employ a self-assembly method to fabricate large-scale microsphere arrays that can be transferred to any desired positions. Additionally, we find that many proposed theories are insufficient to explain the observed properties; including the achieved superresolution, the anomalous effect of immersion, and the size-dependent magnification. These unusual properties demonstrate that half-immersed microspheres are not ordinary solid immersion lenses and new mechanisms must be incorporated to explain their unusual imaging properties.

2. Methods

We employed SiO$_2$ ($n = 1.54$), sapphire (Al$_2$O$_3$, $n = 1.74$), and barium titanate glass (BTG, $n\sim 1.9$-2.1) microspheres with diameters 5~100$\mu$m as the starting materials for making the portable microlens array. The microspheres were assembled on a copper grid following the self-assembly method driven by capillary force described earlier [22]. The processes are shown in Fig. 1(a). First, the microspheres were dispersed into water or alcohol filled in a chamber connected to a syringe. Second, patterned copper grids exhibiting rectangular holes were glued on a supporting washer and fixed on the wall of the chamber. As the surface of the liquid moved, the capillary force dragged the microspheres across the copper grids. The copper grids served as physical traps for the microspheres when gradually pumping out the liquid. By adjusting the pumping speed and the tilt angle of the copper grid, we can routinely achieve regular patterns of microspheres with sizes approaching millimeters, as shown in Fig. 1(b). Although microspheres alone have been reported to achieve superresolution, other studies suggest that half-immersed microspheres yield better contrast and superior image qualities [4,8]. To resolve the controversy, we have systematically studied both configurations. Thus, the self-assembled microspheres were then either immersed in various liquids or spin-coated with poly(methyl methacrylate) (PMMA, $n = 1.49$). The resulting geometry of a PMMA-coated sapphire microsphere is shown in Fig. 1(c). Note that the whole structure now should be regarded as a new lens due to the similar $n$ [4,8]. Then the microlens arrays can be fixed to a manipulator and be transferred to any desired positions under a microscope for further optical characterizations.
We employed blue-ray DVDs and designed metal structures to characterize the images. When imaging a blue-ray DVD, the protection layers of the blue-ray DVD samples were peeled off so that the microlenses could attach to the grating structures. The designed metal structures were fabricated using standard electron-beam lithography followed by 50/5nm thick gold/chrome deposition on a glass substrate and lift-off process. The optical images were inspected under an Olympus microscope equipped with a 50X objective (NA = 0.6), illuminated by a He-Ne laser (\(\lambda = 632.8\)nm) or a white light source. In some experiments, we also employed a home-built confocal microscope to characterize the resolution capabilities.

We first demonstrate that the microspheres are capable of resolving subwavelength gratings without severe distortion. Figures 1(e) and 1(f) show that the subwavelength gratings shown in Fig. 1(d) can be clearly resolved even when imaging at slant angles of 20 and 40 degrees, respectively, demonstrating their capabilities of imaging at wide angles.

3. Results and discussions

Figures 2(a)-2(c) show the blue-ray DVD patterns imaged under PMMA-coated microspheres of different diameters. The blue-ray DVD patterns consist of 200nm-wide aluminum lines separated by 100nm gaps. From the visibility (visibility = \((I_{\text{max}}-I_{\text{min}})/(I_{\text{max}} + I_{\text{min}}))\), where \(I_{\text{max}}\) and \(I_{\text{min}}\) are respectively the maximum intensity and minimum intensity of the selected region of a grating) of the images, we can deduce the point-spread-function (PSF) and its full-width-at-half-maximum (FWHM). For comparison, the Rayleigh criterion for a solid immersion lens is 0.61\(\lambda/n\) (here we have assumed \(NA = n\) for an idealized solid immersion lens), which corresponds to visibility = 0.053–0.105 for \(n = 1.5–1.74\). Figures 2(a)-2(c) demonstrate that the visibilities can respectively reach 0.29, 0.18, and 0.15 for SiO\(_2\) microspheres of 12µm, 39µm, and 89µm in diameter respectively half-immersed in PMMA. The results considerably exceed the diffraction-limited visibility of an idealized solid immersion lens made of SiO\(_2\).
Thus superresolution is unambiguously achieved. From our experiment, the best resolution is $\lambda/4.25$ for PMMA-coated microspheres when imaged under an ordinary microscope.

To optimize the experimental conditions, we have transferred the microspheres to operate under a confocal microscope. Figure 2(d) shows an SEM image of a designed structure consisting of unparalleled two gold lines of 50nm in width on a glass substrate. We also make labels at the side of the gold lines to denote their separations so that the optical resolution can be determined in situ, as shown in Fig. 2(e). The image of Fig. 2(f) clearly resolve the two gold lines separated by 90nm under a confocal microscope at $\lambda = 634$nm laser illumination. After deconvolution, we find the FWHM of the PSF is 99nm, we thus unambiguously determine that the resolution of the PMMA-coated microsphere reaches $\lambda/6.4$. However, we admit that we could not reproduce the previously claimed $\lambda/17$ superresolution based on our confocal imaging [7].

The achieved superresolution and the unusual effect of immersion indicate that the imaging properties of microspheres do not follow the conventional concept of solid immersion lenses. Based on numerical simulations, some previous works proposed a photonic nanojet forming at the focal point of a microsphere to be responsible for the superresolution [3,16,23–25]. However, numerical simulations and experimental studies found the width of the nanojet to be $\sim \lambda/2$, disagreeing with our and other results [17,18]. Later, it was suggested that resonant nanojet would appear at a narrower width $\sim \lambda/3$ [20,21], but it is still insufficient to explain the observed superresolution.

The theory of nanojet has been used for explaining the effect of immersion as well [11]. It is predicted that the resolution is a function of the ratio between the refractive index of the microsphere ($n$) and that of the immersion media ($n_m$), and it can be optimized when $n/n_m =$
1.55 [11]. Thus, the validity of the theory can be investigated using different microspheres immersed in various liquids and compare the changes of resolution before and after the liquid dries (for liquids that do not evaporate easily, the experimental procedure is reversed; that is, the microspheres were first inspected in air and then immersed in the liquids). Experimentally, we have immersed different microspheres in water ($n_m = 1.33$), isopropyl alcohol (IPA, $n_m = 1.36$), acetone ($n_m = 1.38$), PMMA ($n_m = 1.49$), and microscope immersion oil ($n_m = 1.515$). Figure 3 shows the FWHM of PSF (normalized to peak intensity wavelength $\lambda = 600$nm of the incident white light source) versus $n/n_m$. The prediction of the nanojet theory is also shown for comparison.

We now divide the anomalous immersion effects in Fig. 3 into two parts. For $n/n_m < 1.3$, we find that the resolution is critically dependent on whether the microspheres are half-immersed or fully-immersed. In fact, except for BTG microspheres, all other microspheres cannot resolve the blue-ray DVD pattern when they are in air or fully-immersed, suggesting that the resolution is worse than $0.61\lambda$. Although one might attempt to attribute the reduction of resolution to acceptance angles of the microlens, simulations on these geometries show that the decrease of the acceptance angle from half-immersed ($45^\circ$) to fully-immersed ($40^\circ$) is too small to account for the effect [4]. Previous works have instead attributed the result to the image plane of a fully-immersed microsphere being far away from the sample so it cannot be captured by the microscope objective [6]. For $n/n_m < 1.3$, it is nevertheless anomalous that the resolution of microspheres half-immersed in different media is much better than the theoretical prediction.

For $n/n_m > 1.3$, we find that the resolution is material dependent. For example, distinct resolution is found when fully immersing BTG microspheres in oil and in PMMA even though their $n_m$’s are similar ($n/n_m = 1.33$). Another example shows that when BTG microspheres are fully immersed in water, they always yield better resolution than other material combinations even if their $n/n_m$’s are nearly identical ($n/n_m \approx 1.55$). Moreover, whenever the immersion liquid dries, the resolution becomes poor than $0.61\lambda$, disagreeing with the theoretical prediction.

The results of Fig. 3 also suggest that BTG microspheres are notable exceptions. For example, half-immersed SiO$_2$ and sapphire microspheres always give superior resolution than those of fully-immersed configurations, but the conclusion does not apply to BTG
microspheres. Furthermore, only the BTG microspheres display the anomalous material dependent resolution mentioned above.

As for magnification \((M)\), ray optics gives \(M \sim n/[n_m(2-n/n_m)]\) for a spherical lens \((n)\) fully-immersed in a media \((n_m)\). Thus, \(M = 1.06, 1.32,\) and \(2.25\) for SiO\(_2\)/PMMA, sapphire/PMMA, and BTG/oil, respectively. We have systematically studied the \(M\)'s of different microspheres and plotted the result in Fig. 4. It can be seen that for large BTG microspheres fully-immersed in oil, their \(M\) agrees with the result of ray optics. However, their \(M\)'s start to deviate from the predicted value when the diameters are smaller than 15\(\mu m\). Large deviation can be also found in SiO\(_2\)/PMMA whose \(M = 3.2-4.1\), more than three times larger than the prediction based on ray optics. Interestingly, the most pronounced deviation is found in sapphire/PMMA, in which not only the \(M\)'s but also the diameter dependence displays entirely different characteristics.

Previous works have noticed the unusual diameter dependence of \(M\) and proposed to utilize intensity enhancement at the focal point to empirically explain the effect [9]. Based on numerical simulations, it is found that \(M\) is a linear function of diameter, similar to the behavior observed in sapphire/PMMA in Fig. 4. However, the empirical model has not fully elucidated the complex behavior shown in Fig. 4. In particular, while both \(n\) and \(n/n_m\) increases from the cases of SiO\(_2\)/PMMA, sapphire/PMMA, to BTG/oil, \(M\)'s instead display nontrivial dependencies and it certainly cannot be a monotonically increasing function of \(n\) or \(n/n_m\).

![Fig. 4. Magnification vs. diameters of various microspheres that are either half-immersed (half-filled symbols) or fully-immersed (solid symbols) in different media. The lines are guides to the eyes.](image)

The above results suggest that we have not reached full understandings on the superresolution mechanism of the microspheres. Although the theory of nanojets is based on full-wave simulations and there are believes on their accuracies, it nevertheless disagrees with many experimental observations. It is possible that previous simulations ignored the dispersion relations of dielectric materials and the contributions of surface waves at interfaces were overlooked. Recently, Regan et al suggested that surface waves between two dissimilar dielectrics to be the mechanism for superresolution [26]. They demonstrated that evanescent waves can couple to the surface waves and then transmit to far field whenever the condition for momentum match is satisfied between the surface and the air/substrate [26]. In Regan et al’s experiment, they deposited a PMMA layer doped with Rhodamine-6G and the fluorescent image showed that the surface waves indeed carry high spatial resolution.
information (\(\lambda/5\)) of the sample to far field at angles larger than critical angles determined by superstrate/substrate [26]. However, their planar geometry is different from the microspheres discussed here and transmitting the information to far field was done via fluorescence rather than direct imaging of the reflected light. Although Regan et al’s explanation may not directly apply to our results and there are still unknowns about the coupling processes, it nevertheless emphasizes the important contribution of surface waves.

In summary, we have applied rigorous deconvolution analyses and determine that the best spatial resolution of a half-immersed microsphere can reach \(\lambda/6.4\). We also employ a low-cost, self-assembly method to fabricate large-scale microsphere arrays that can be transferred to any desired positions. Experimentally, we reveal many interesting optical properties of microspheres; including the achieved superresolution, the anomalous effects of immersion, and the unusual size-dependent magnification. It is suggested that new mechanisms, such as surface waves, must be considered to understand their imaging properties.

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