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**Title:** Optical nano-imaging via microsphere compound lenses working in non-contact mode

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Optical nano-imaging via microsphere compound lenses working in non-contact mode: supplemental document

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1. The imaging schematics of MCL working in different imaging configurations

(a) virtual-virtual  (b) real-real  (c) real-virtual

Fig. S1. The imaging schematics of MCL working in (a) virtual-virtual (VV), (b) real-real (RR) and (c) real-virtual (RV) imaging configurations.

2. Derivation of image distance, magnification and conditions for virtual-real imaging configuration

The schematic of MCL for nano-imaging is presented in Fig. S2. According to the geometric optics theory [1, 2], the imaging distance (position) and magnification of a single microsphere can be calculated by Eq. (S1) and Eq. (S2), respectively.

\[
p = \frac{(2n_0-n_i) r_l - 2n_0 r^2}{2(n_i-n_0) l + 2n_0 r - n_1 r} + 2r, \quad \text{(S1)}
\]

\[
\beta = \frac{n_1 r}{2l(n_i-n_0) + r(2n_0-n_i)}. \quad \text{(S2)}
\]
The derivation of Eq. (S1) and Eq. (S2) are based on the relationship between object and image distance for a single spherical surface [1]. Where \( l \) and \( l' \) are object distance and image distance, respectively. The origin point used for measuring the object distance and image distance is same, i.e. the pole of the bottom spherical surface of the microsphere. If object or image locate at the bottom side of the origin point, \( l \) or \( l' \) takes a negative value. Otherwise, they take a positive value. So, in equation (S1) and (S2), the object distance \( l \) should take a negative value for calculation. \( \beta \) is the magnification coefficient. \( r \) is the radius of microsphere. \( n_0 \) and \( n_1 \) are the refractive index of environment and microsphere, respectively.

In the MCL, the image distance and magnification of the bottom microsphere can be calculated with Eq. (S1) and Eq. (S2). The upper microsphere takes the image produced by bottom microsphere as the sample and magnified it again. The origin point used for measuring the object distance and image distance of the upper microsphere is the pole \( P_2 \) of the bottom spherical surface of upper microsphere. Due to the change of origins in the calculations for bottom and upper microspheres, the object distance for the upper microsphere is the image distance of the bottom microsphere plus the gap between two microspheres and minus diameter of the bottom microsphere. The calculation of image distance (position) and magnification for upper microsphere can also reuse Eq. (S1) and Eq. (S2) by replacing the value of object distance accordingly. It is an iterative process. So, the image distance \( l'_{\text{MCL}} \) and magnification \( \beta_{\text{MCL}} \) of a MCL can be calculated by Eq. (S3) and Eq. (S4) as following:

\[
\begin{align*}
\frac{l'}{l} &= \left( \frac{2n_0 - n_1}{n_1} \right) \frac{r}{l - 2n_0 r^2} + 2r_i \\
\frac{l'_{\text{MCL}}}{l} &= \left( \frac{2n_0 - n_1}{n_1} \right) \left( \frac{r}{l' + g - 2r_i} \right) - 2n_0 r_i^2 + 2r_2 \\
\beta_1 &= \frac{n_1 r_i}{2l(n_1 - n_0) + r_i (2n_0 - n_1)} \\
\beta_2 &= \frac{n_1 r_i}{2(n_2 - n_1)(l' + g - 2r_i) + r_i (2n_0 - n_1)} \\
\beta_{\text{MCL}} &= \beta_1 \times \beta_2 
\end{align*}
\]  

(S3)

To note, if a distance locates at the bottom side of \( P_2 \), it is a negative value. Otherwise, it is a positive value. So, in equation (S3), the gap \( g \) should take a negative value for calculation.

Besides, if we obtained a positive \( l'_{\text{MCL}} \), it means the image produced by the MCL locate at the upper side of \( P_2 \). Otherwise, the image produced by the MCL locate at the bottom side of \( P_2 \).

When the special case that a MCL is put near the sample surface (\( l \approx 0 \)) and the two microspheres kiss with each other (\( g=0 \)) is considered, based on the above iterative processes, we can obtain a simplified magnification coefficient \( \beta_{\text{MCL}} \) of the MCL:

\[
\beta_{\text{MCL}} = \frac{1}{2} \left( \frac{n_1}{r_1} \right) \left( \frac{n_2}{r_2} \right) \left( \frac{n_0}{r_0} \right) \\
\]  

(S4)

Where \( r_1 \) and \( r_2 \) are the radius of bottom and upper microsphere, respectively. \( n_0, n_1 \) and \( n_2 \) are the refractive index of environment, bottom microsphere, and upper microsphere, respectively. When the upper microsphere works near the boundary between real and virtual imaging manner, \( \beta_{\text{MCL}} \to \infty \). The radius ratio of two microspheres should satisfy:
When the magnification $\beta_{\text{MCL}}$ of the MCL is 1, the radius ratio of two microspheres should satisfy:

$$\frac{r_2}{r_1} = \frac{4n_0(n_2-n_0)}{(2n_0-n_1)(2n_0-n_2)} = k_1, \quad (S6)$$

Hence, the condition for the MCL working in virtual-real (VR) imaging configurations and remaining magnifying function is:

$$k_1 > d_2/d_1 > k_2. \quad (S8)$$

Where $k_1$ and $k_2$ are the two boundaries. Significantly, if we substitute the refractive indices of material with effective refractive index of microsphere in all the above equations, we can calculate the related parameters with higher precision.

3. The differences in the focal lengths of microsphere simulated by 3D and 2D FDTD model

The focal length of microsphere simulated by 3D and 2D FDTD models are slightly different. To check the difference, we have simulated focal lengths of five silica microspheres (diameter 10 μm – 18 μm) at 405 nm wavelength using both 3D and 2D FDTD models. The results are shown in the Fig. S3. It can be found that the relative ratio $\Delta f_r$ ($\Delta f_r = |f_{2D} - f_{3D}|/f_{2D}$) of change in the focal length obtained by 2D and 3D FDTD models is very small (<0.044) within the simulation range. Besides, the overall variation trend is that $\Delta f_r$ reduces as the increase of microsphere diameter. From this study, we predict that the difference between 2D and 3D FDTD models should have limited effects on our conclusion. In addition, the 3D model always requires a significantly larger running memory. For example, the RAM of the working station used for simulation in this work is 36 GB, which ultimately can support the full-wave simulation for a single ~18 μm silica microsphere with 3D FDTD model (meshing accuracy is set as default level 2 in FDTD software). Although the simulation for microsphere by 3D model should be more precise, limited by the simulation resources and for consistency, the 2D FDTD models are employed to simulate the focal length of all 10 μm - 120 μm microspheres in this work.

Actually, simulating the focal length of microsphere with 2D model is widely adopted in literatures. Although the focal lengths of microsphere obtained by 2D model are not very accurate, without loss of generality, it can still reflect the fundamental trend of changes. In this
work, the simulations for silica microsphere and MCL are mainly utilized as an example to demonstrate the effectiveness of the concept “effective refractive index”. Since both the calculation of effective refractive index and full-wave imaging simulation are based on 2D FDTD model, considering the slight difference between 2D and 3D models, their results presented in Fig. 2 will not have significant changes if 3D model simulation is used. In comparison with the ray tracing results in the same figure, such small discrepancies do not affect our analysis and conclusion.

4. Predictions of imaging configurations with effective refractive index and refractive index of material

To demonstrate the effectiveness of the effective refractive index on predicting imaging configurations, an MCL consisting of 10 μm and 68 μm silica microspheres is designed to image three dipole sources by simulation. The results of 2D ray tracing simulation with refractive index of silica and effective refractive index are presented in Fig. S4. (a) and (b) respectively. The results with refractive index of silica and effective refractive index predict that the MCL should works in VV imaging configuration and VR imaging configuration, respectively. The 2D full-wave simulation result with refractive index of silica, as shown in Fig. S4. (c), confirms that this MCL should work in VR imaging configuration. These results demonstrate that the geometric optics theory with effective refractive index can provide accurate imaging configuration prediction, whereas the geometric optics theory with refractive index of material exhibits large deviation.

Fig. S4. The images of three-point sources simulated by ray tracing with (a) refractive index of silica, (b) effective refractive index of silica microspheres, and by (c) FDTD full-wave simulation with refractive index of silica. The MCL used in this case consists of 10 μm and 68 μm silica microsphere. P represents optical power.
Some key software settings for ray tracing simulation in COMSOL include: a. mesh: default “finer” accuracy and b. maximum length of each step: 80 nm. The mesh accuracy for full-wave 2D FDTD simulation are default level 2 accuracy in Lumerical FDTD software. The hardware settings of the work station used in the work are shown below: a. Processor: Intel(R) Xeon(R) CPU E5-2697 v2 @ 2.70 GHz, 2.70 GHz (2 processors); b. RAM: 36 GB; c. Hard Disk: 3 TB.

5. The SEM image of Blu-ray disc

The minimum feature size and period of the Blu-ray disc used in experiments are 100 nm and 320 nm, respectively. The SEM image of Blu-ray disc is presented in Fig. S5.

![SEM image of Blu-ray disc](image.png)

**Fig. S5.** The SEM image of the Blu-ray disc used in experiment. The minimum feature size is ~100 nm and the period is ~320 nm.

6. Experimental procedures to control and validate the gap distance between the two microspheres

The schematic of experimental procedures to control and validate the gap distance between the two microspheres are shown in Fig. S6. The two microspheres in MCL are mounted on two holders which are connected to and controlled by two separate nano-stages. The two nano-stages together with the sample are fixed on a microscope sample stage which possesses a high movement resolution (~100 nm). The real-time z coordinate of the sample stage surface is displayed in the control software. In the first step, the two microspheres are kissed with each other. The equator of the upper microsphere is moved up to the imaging plane of the objective lens together with the sample stage. Then, the z coordinate of the sample stage is recorded. In the second step, an isolated upward movement of the upper microsphere to a certain distance is induced by the connected nano-stage. In the third step, the upper microsphere is moved down with the sample stage so that its equator is relocated at the imaging plane of the objective lens. The current z coordinate of the sample stage is recorded, and it minus the z coordinate recorded in step 1 is equal to the gap distance $g_1$ between the microspheres after the first movement of nano-stage connected to the upper microsphere. Steps 4 and 5 are just repeated procedures described in steps 2 and 3, and then we can obtain the final gap distance after the series of movements between microspheres. Since the movement resolution of the sample stage is ~100 nm and the depth of field of the microscope with $100 \times$ objective lens is ~800 nm, the measurement accuracy for the gap distance is ~800 nm. According to experimental results shown in Fig 4(c) in the manuscript, the magnification does not change significantly with a ~800 nm variation of gap distance. So, the ~800 nm measurement accuracy for gap distance is good enough to verify the relationship between magnification and gap distance of microspheres.

From the experimental procedures, the feedback mechanism for controlling and validating the gap distance is using a close-loop sample stage with accurate and quantified positioning to measure the equivalent gap distance with the assistance of optical microscopic imaging when the upper microsphere settles down after each movement. The gap distance measured by the feedback mechanism can be compared with the given movement distance of the nano-stage for further validation.
7. The effect of the working distance of bottom microsphere on magnification of microsphere compound lens

The working distance of the bottom microsphere in experiment is very short and difficult to be accurately measured by simple methods, including the one described in section 6 for gap distance measurement. So, in the experiment, we estimated that the microsphere compound lens working in non-contact mode through some indirect evidences, like the microsphere can smoothly move above sample for scanning mode imaging. Since the working distance of the bottom microsphere in experiment is quite short, the effect of working distance on magnification calculation can be almost neglected. As an example, the magnification of an MCL consisting of 23 μm and 110 μm silica microspheres when its working distance varies from 0 to 500 nm is calculated based on the modified theory and shown in Fig. S7. The magnification decreases as the increase of working distance. However, the variation within this range is very small. Therefore, considering the working distance is quite short, its effect on calculations of magnification of the compound lens system is insignificant.

8. Calculations of focal lengths, effective refractive indices, and diameter ratios for VR imaging configuration.

The focal lengths and effective refractive indices of barium titanate glass microspheres (n~2.2) in oil environment (n~1.5) are shown in Fig. S8. (a) and (b). The diameter ratio for VR imaging configuration when MCL is compose of barium titanate glass microspheres and works in oil environment is presented in Fig. S8. (c). In the experiment, an MCL consisting of 30 μm (bottom) and 80 μm (upper) barium titanate glass microsphere working in oil environment is
utilized. Since the diameter ratio is ~2.67, it works in VR imaging configuration according to Fig. S8. (c).

Fig. S8. The focal length (a) and the refractive index (b) versus the diameter of barium titanate glass microsphere (n~2.2) at 405 nm wavelength. The microspheres are immersed in oil environment (n~1.5). The blue and red line in (a) is calculated with geometric optics formula and obtained from FDTD simulation, respectively. The blue and red lines in (b) represent the refractive index of barium titanate glass and effective refractive index of barium titanate glass microsphere, respectively. (c) The diameter ratio of two microspheres for VR imaging configuration calculated with refractive index of barium titanate glass material (region B) and effective refractive index of barium titanate glass microsphere (region A).

References
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