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In-Situ Characterization of Three-Dimensional Optical Matters by Light Diffraction *

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Three-dimensional optical matters are created by combining the single beam optical trapping with the conventional Z-scan technique. Dynamic light diffraction is employed to evaluate the structure and quality of the optical matter formed at the optimum trapping power. The lattice constant of the optical matter is extracted based on the Bragg and Snell laws, showing that polystyrene spheres are nearly close-packed in the optical matter, confirmed by comparing the diffraction pattern of the optical matter with that of a colloidal photonic crystal fabricated by the self-assembled technique. The relatively broad diffraction peaks observed in the optical matter indicate that the density of disorders in it is higher than that in the photonic crystal. It is suggested that the optical matter possesses a random close-packed structure rather than a face centered cubic one.

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Single beam optical trapping, as first reported by Ashkin *et al* in 1986, allows one to trap and manipulate micro- and nanoparticles in a simple and effective way.[1] This technique, which is now called optical tweezers, has found extensive applications in various fields.[2] In the last two decades, research in this field has progressed from the trapping of single particles to the manipulation of a large number of particles with the hope of building ordered structures. So far, the most popular technique used to trap and manipulate multiple particles is based on holographic optics.[3] The ordered structures created and held by photons are referred to as optical matters.[4,5] They are distinct from photonic crystals (PCs),[6] which have been studied extensively in the last two decades, with their dynamic and reversible features. However, the disadvantage of the holographic technique is the complexity of experimental arrangement. Therefore, a simple and efficient way to create three-dimensional (3D) optical matters with large volumes is highly desirable.

Recently, we proposed a simple method to create 3D optical matters based on the combination of the single beam optical trapping and the conventional Z-scan technique.[7] In addition, optical matters with large volumes were also achieved by optically trapping polystyrene (PS) spheres contained in a capillary and well-reproduced optical switching with a large extinction ratio (about 20 dB) was realized.[8] In both the cases, objective lenses with small magnifications (5× or 10×) were used in order to avoid the strong scattering force in the forward direction. This choice makes it difficult to identify the structure and quality of the formed optical matters. However, many physical properties of the optical matters remain to be unveiled, such as the lattice constant, the quality and the structure.

Modern microscopy allows 3D imaging of materials, mainly giving information on local structure and ordering. In comparison, light diffraction or scattering techniques are more appropriate for the study of soft materials with long-range ordering.[9−15] Among them, Bragg scattering has been adopted to characterize the quality of ordered soft materials grown in a microgravity environment.[16] Optical matters, as one kind of soft materials, are generated in a gradient optical field. Therefore, the nucleation and growth processes of optical matters are expected to be much different. Due to the difficulties mentioned above, we have to rely on light scattering to reveal the physical properties of the formed optical matters. In this Letter, we report on the in-situ characterization of 3D optical matters generated in the Z-scan-based optical trapping by use of light diffraction.

The dynamic light diffraction conducted for optical matters is schematically shown in Fig. 1. The experimental setup is quite similar to that for the Z-scan-based optical trapping.[7] The 532 nm light from a solid state laser was used not only for the trapping of PS spheres but also as the light source for the diffraction experiments. It was focused by a 5× objective lens onto the sample cell. The 50-μm thick sample cell contained the aqueous solution of 1.9 μm PS spheres (Duke Scientific) with a concentration of 10 wt.%. The diffraction pattern of the trap region

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was projected on a white screen. When measuring the full widths of the diffraction peaks, a 20× objective lens was used to focus the diffraction pattern into a charge coupled device (CCD) whose size is only $6.8 \times 5.6\,\mathrm{mm}^2$. In this case, the sample cell, the 20× objective lens and the CCD were fixed on the same moving stage. The scanning processes were performed in a region of $z \in (-5.7625, 0)\,\mathrm{mm}$ with a scanning speed of 191 μm/s. Correspondingly, the beam diameter was varied from about 2 mm to about 10 μm.

Once the trapping power P_t exceeded a threshold (about 15 mW in this case), a diffraction pattern composed of regularly distributed Bragg diffraction spots began to appear on the screen when the sample cell approached the focus point (see Fig. 2), indicating the formation of an optical matter.[7] At first glance, the diffraction pattern of an optical matter looks like the one observed previously for a colloidal PC illuminated by an incident light along the [111] direction. In general, a colloidal PC formed by the self-assembly of PS spheres usually possesses a close-packed structure in which hexagonal lattices of PS spheres are stacked layer by layer in different sequences, forming a face centered cubic (fcc), a twin face centered cubic ($tfcc$), a hexagonal close-packed (hcp), or a random close-packed (rcp) structure etc.[10–14]

When the diffraction of a colloidal PC is concerned, the colloidal PC is usually considered as the stack of 2D gratings and the diffraction pattern is obtained by the superposition of the diffractions from all 2D gratings in a single scattering configuration.[11,14] If a coordinate system is established so that the hexagonal lattices (or 2D gratings) are parallel to the xy plane, the primitive lattice vectors for the hexagonal lattices are generally chosen as $\boldsymbol{a}_1 = d\boldsymbol{x}$ and $\boldsymbol{a}_2 = d(\boldsymbol{x} + \sqrt{3}\boldsymbol{y})/2$, where d is the lattice constant of the hexagonal lattices which is equal to the diameter of spheres in a close-packed structure. Then, the primitive lattice vectors in the reciprocal space are derived to be $\boldsymbol{b}_1 = 4\pi\boldsymbol{y}/(\sqrt{3}d)$ and $\boldsymbol{b}_2 = 2\pi(\boldsymbol{y}/\sqrt{3} - \boldsymbol{x})/d$ and the corresponding lattice vector can be expressed as $g = pb_1 + qb_2$, where (p, q) is a pair of integers.[13] To do so, the six first-order diffraction spots observed in the diffraction pattern can be indexed as $(p, q) = (1, 0), (1, -1), (0, -1), (-1, 0), (-1, 1)$, and (0,1). If the wavelength of the incident light is fixed, the relationship between the diffraction angle θ_w and the lattice constant of the hexagonal lattice d is deduced to be

$$\sin\theta_w = 2\lambda/(\sqrt{3}d), \tag{1}$$

where λ and θ_w represent the wavelength and diffraction angle of light in water. As schematically shown in Fig. 1(b), the diffracted light suffers from refraction at the rear wall of the sample cell. Since the thickness of the wall is quite small, the refraction of the diffracted light can be considered to occur at the interface between water and air. Thus, the diffraction angle in water and in air, i.e. θ_w and θ_a can be related by the Snell law

$$n\sin\theta_w = \sin\theta_a, \tag{2}$$

where $n = 1.33$ is the refractive index of water. From Eqs. (1) and (2), the lattice constant of the hexagonal lattices can be derived as

$$d = 2\lambda_0/(\sqrt{3}\sin\theta_a), \tag{3}$$

where $\lambda_0 = 532\,\mathrm{nm}$ is the wavelength of light in vacuum. In experiments, we fixed the distance between the screen and the focus point of the objective lens, where the scanning processes were stopped. By monitoring the diffraction patterns, it was found that the formed optical matters could live for a long time– several minutes. Thus, the separation between the zero- and first-order diffraction spots was easily determined. A typical diffraction pattern of the optical matter obtained at the optimum trapping power of about 100 mW is shown in Fig. 2. After examining many diffraction patterns, the average value of $\sin\theta_a$ was derived to be 0.32. Thus, the lattice constant of the hexagonal lattices is deduced to be $d = 1.9173\,\mu\mathrm{m}$. This value is slightly larger than the diameter of PS spheres, indicating that the optical matter is formed by nearly close-packed PS spheres.

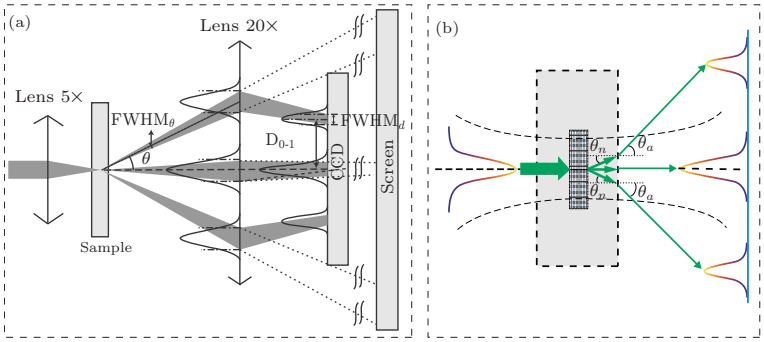

Fig. 1. (a) Experimental setup for dynamic light diffraction used to characterize the three-dimensional optical matters created in Z-scan-based optical trapping. (b) Schematic showing the diffraction of light by an optical matter created in water and the refraction of the diffracted light at the rear wall of the sample cell which is approximated as an interface between water and air.

Fig. 2. Diffraction pattern of the optical matter obtained at the optimum trapping power of about 100 mW. It was projected on a screen placed several centimeters away from the sample cell.

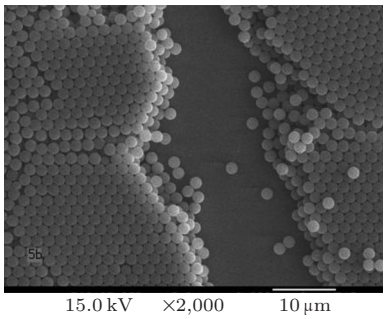

Fig. 3. SEM image of the colloidal PC fabricated by the pressure controlled isothermal heating vertical deposition technique.

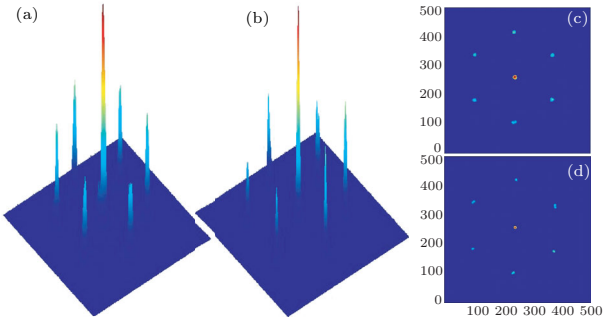

Fig. 4. Three- and two-dimensional presentations of the diffraction patterns for the optical matter ((a) and (c)) and the PC ((b) and (d)) recorded by the CCD.

Table 1. Parameters derived from the diffraction pattern of the optical matter obtained at the optimum trapping power of ~ 100 mW. The definitions of these parameters are D_{0-1}: the separations between the zero- and first-order diffraction peaks; FWHM$_d$ and FWHM$_\theta$: the full width of the diffraction peaks represented in length and in angle; I and P: the relative intensity and power distributions among the diffraction spots. The values for the zero-order diffraction spot are used for normalization. 1 pixel = 6.7 µm.

	Zero-order	First-order diffraction peaks					
D_{0-1} (pixel)	0	154.052	151.268	160.178	150.841	163.615	163.110
FWHM$_d$ (pixel)	14.842	12.666	13.541	12.048	13.588	14.002	16.077
FWHM$_\theta$ (rad)	0.01620	0.01267	0.01380	0.01159	0.01389	0.01320	0.01521
I	1	0.52563	0.50496	0.49609	0.49510	0.40944	0.35324
P	1	0.34143	0.40648	0.31116	0.35373	0.32192	0.38183

Table 2. Parameters derived from the diffraction pattern of the PC fabricated by the PCIHVP technique. The definitions of these parameters are D_{0-1}: the separations between the zero- and first-order diffraction peaks; FWHM$_d$ and FWHM$_\theta$: the full width of the diffraction peaks represented in length and in angle; I and P: the relative intensity and power distributions among the diffraction spots. The values for the zero-order diffraction spot are used for normalization. 1 pixel = 6.7 µm.

	Zero-order	First-order diffraction peaks					
D_{0-1} (pixel)	0	157.953	155.116	168.048	160.378	164.128	172.186
FWHM$_d$ (pixel)	10.464	9.707	10.218	9.027	13.062	9.508	12.616
FWHM$_\theta$ (rad)	0.01102	0.00946	0.01015	0.00827	0.01256	0.00892	0.01129
I	1	0.53624	0.50519	0.44327	0.38892	0.34090	0.31132
P	1	0.51517	0.53143	0.46279	0.56367	0.37195	0.48695

Having known that PS spheres in the optical matter are close-packed in the transverse direction, it is believed that the close-packed form also occurs in the longitudinal direction. In principle, a diffraction pattern can be observed even though there are only a few layers of PS spheres in the direction of the trapping light. We have investigated the dependence of the quality of the formed optical matters on the trapping power by monitoring the diffraction pattern of the trap region.[17] It is found that the quality of the optical matters is clearly reflected in the full widths of the diffraction peaks. At high trapping powers, a significant broadening of the diffraction peaks is observed, implying a dramatic degradation in the quality of the optical matters caused by the strong scattering force. Similar broadening of the diffraction peaks has been found in PCs in the presence of disorder.[18] It has been found that the best optical matter is obtained at an optimum trapping power of about 100 mW. In order to gain a deep insight into the crystal quality of the optical matter, we have compared its diffraction pattern with that of a self-assembled PC. The PC was fabricated by using the pressure controlled isothermal heating vertical deposition (PCIHVD) technique from a solution containing 1.9 µm PS spheres with a concentration of 1.0 wt%.[19,20] The scanning electron microscope (SEM) image shown in Fig. 3 indicates that PS spheres in the PC are packed closely with few disorders. The total number of PS layers in the growth direction is four or five. A comparison of the diffraction pattern between the optical matter and the PC is presented in Fig. 4. The 3D presentations of the diffraction patterns of the optical matter and the PC are compared in Figs. 4(a) and 4(b) while the 2D cases are compared in Figs. 4(c) and 4(d). The extracted data for the separations between the zero- and first-order diffraction peaks D_{0-1}, the full width of the diffraction peaks represented in length and in angle, i.e. FWHM$_d$ and FWHM$_\theta$, and the relative intensity I and power P distributions among the diffraction peaks are compared in Tables 1 and 2 for the optical matter and the PC. It can be seen that the separations between the zero- and first-order diffraction peaks in both the samples are quite similar. This implies that the lattice constant in the optical matter is close to that in the PC. Since PS spheres are close-packed in the PC, it is verified that the derivation of the lattice constant of the optical matter presented above is valid although it is immersed in water. A remarkable feature found in Tables 1 and 2 (or in Fig. 4) is that the full widths of the diffraction peaks of the optical matter are broader than those of the PC. Therefore, it is suggested that

the volume density of imperfections in much higher in the optical matter because it is formed by dynamic trapping. Accordingly, more power is diffracted into the first-order diffraction spots in the PC as compared to the optical matter where most power is still concentrated in the zero-order diffraction spot.

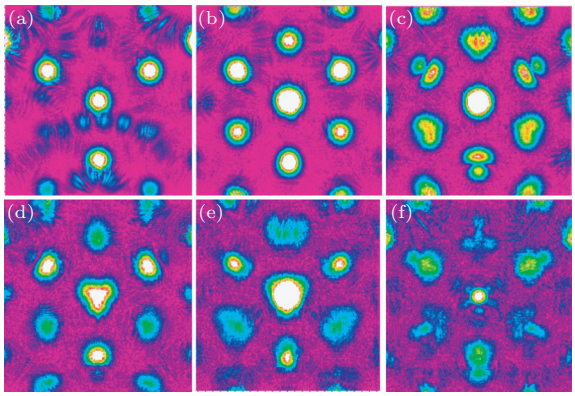

Fig. 5. Evolution of the diffraction pattern with increasing number of layers for a PC possessing a close-packed fcc structure. The number of hexagonal lattices in the longitudinal direction is (a) $N = 3$, (b) $N = 6$, (c) $N = 9$, (d) $N = 12$, (e) $N = 15$, (f) $N = 18$.

In general, a colloidal PC formed by the self-assembled technique usually possesses an fcc structure whose diffraction pattern contains only three first-order diffraction spots.[11] However, this is true only for a perfect fcc structure with an infinite number (or a sufficiently large number) of layers. For an fcc structure with a small number of layers, six diffraction spots are generally observed. In Fig. 5, we present the evolution of the diffraction pattern of a close-packed fcc structure with the increasing number of layers simulated by using a vector beam propagation method.[21] It can be seen that the diffraction pattern exhibits only three first-order diffraction spots when the number of hexagonal lattices is three. When the number of hexagonal lattices is increased to six, six first-order diffraction spots are observed. A further increase of the number of hexagonal lattices leads to the reduction in the intensities of the three diffraction spots and eventually a diffraction pattern with only three first-order diffraction spots is achieved for a sufficiently larger number of hexagonal lattices. It is difficult to derive the number of layers for the optical matter in the direction of light propagation. Based on the simulation results shown in Fig. 5, it is thought that the total number of layers in this direction should be larger than three because six diffraction spots are observed in the diffraction pattern. It was reported that the $tfcc$ or rcp structure might occur in self-assembled PCs.[11–14] In the fabrication of colloidal PCs, it generally takes a long time (e.g. several hours) to obtain the ordered structures. Even in this case, stacking faults are usually found in colloidal PCs and perfect fcc structures are seldom obtained. In our case, the dynamic formation of optical matters occurs within a short time period (less than one minute). Therefore, we intend to believe that the optical matter formed by the Z-scan-based optical trapping possesses a rcp structure.

In summary, we have employed dynamic light diffraction to characterize the 3D optical matters. The lattice constant of the optical matter formed at the optimum trapping power is successfully derived. It is verified that the optical matter is formed by close-packed PS spheres with a rcp structure. Our research work provides a convenient and powerful way to investigate the formation process of soft materials.

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[21] A commercially available software developed by Rsoft Design Group (http://www.rsoftdesign.com) is used for the numerical simulations.