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## Research Article

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# **Orbiting light around the outer surface of dielectric microspheres**

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## **Abstract**

Circulating light in the total internal reflection within dielectric spheres or disks is called the whispering gallery mode (WGM), which by itself is highly sensitive to its surface and capable of detecting viruses and single atomic ions. The detection site of the sensors using WGM is created by the evanescent light from the circulating light inside spheres. On the other hand, there have been no reports of observation or discussion of the light orbiting outside the surface of dielectric microspheres. Here we report light orbiting the outer surface of a dielectric microspheres different from the WGM. We observed anomalously enhanced Raman spectrum at the periphery of 3  $\mu\text{m}$  diameter polystyrene (PS) microspheres on a silicon nitride (SiN) film using Raman microscopy. The wavelength intensity of this enhanced Raman spectrum was accompanied by periodic changes due to interference. These features may lead to the development of high-sensitive sensors and optical devices.

(150 words)

## **Keywords:**

Dielectric microsphere, Confocal Laser Raman microscopy, Anomalously enhanced Raman spectrum, Fluorescence image, Orbiting light, Polystyrene, Silicon nitride film

Dielectric spheres and disks show peculiar properties such as WGM<sup>1-8</sup>, Mie scattering<sup>9-12</sup> and surface enhanced Raman spectrum (SERS)<sup>13-16</sup>. These phenomena are generated by electric and magnetic dipole resonances inside dielectric micro- or nanospheres, disks and cylinders by light irradiation<sup>17-19</sup>. The incident light on a dielectric microsphere circulates at the internal surface of the sphere with total reflection<sup>17-19</sup>. At this time, the incident light is drastically amplified and exhibits a high-quality factor WGM<sup>5-7</sup>. The surface of the WGM devices generates evanescent light due to the internally orbiting light and functions as an extremely sensitive sensor for viruses<sup>4</sup>, proteins<sup>2</sup> and single atomic ions<sup>3</sup>. In addition, WGM of the microspheres and disks can be applied to sensors<sup>1-4</sup>, resonators<sup>5-8</sup>, laser<sup>20-24</sup>, quantum and nonlinear optics<sup>25-27</sup>.

Here, we demonstrate the existence of orbiting light on the outside surface of the dielectric microspheres for the first time to our knowledge. This orbiting light was induced when the sphere was attached to a SiN film but not detected on other substrates such as a slide glass or a SiN film with Si frame. Moreover, the orbiting light strongly influenced by the thickness of SiN film and the sphere diameter. This external circulating light can be observed under a confocal laser Raman microscopy and

conventional fluorescence microscopy. This orbiting light on the microsphere is extremely sensitive to the surface condition of the spheres and the contact site of the SiN film, and is expected to be used for highly sensitive compact sensors, antennae and optical quantum elements.

## **Results**

### *Anomalous Raman spectrum of dielectric microspheres on a SiN film*

PS microspheres attached on a SiN film were observed under a confocal laser Raman microscope (Fig. 1a). The 3  $\mu\text{m}$  diameter spheres were dispersed on a SiN film of 50 nm thickness supported by a Si frame with a square window; the SiN film over the window was not directly contacted by the Si frame (Fig. 1b and c). We compared Raman spectra of PS spheres on the SiN film with and without direct contact of the Si frame (Fig. 1d–f). At the centre of the sphere on the SiN film without contact with the Si frame, the Raman spectrum showed a PS peak of  $1,006\text{ cm}^{-1}$  (Fig. 1d red line). Notably, we observed anomalously enhanced Raman spectra with a periodic amplitude change at the periphery of spheres (Fig. 1d blue line). Furthermore, the PS peak at  $1,006\text{ cm}^{-1}$  was very small, demonstrating that the laser did not irradiate the inside of the spheres directly (Fig. 1e). Moreover, this result suggests that the incident light does not

circulate inside the spheres. When the light circulates inside the spheres by WGM, a large PS peak should be observed<sup>16</sup>. For PS spheres attached on the SiN film contacted by a Si frame, Raman peaks of Si at  $525\text{ cm}^{-1}$  and of PS at  $1,006\text{ cm}^{-1}$  were observed at the centre and periphery, but without any periodic anomalous Raman spectra (Fig. 1f). The effect of Si underneath the SiN film should be very small if the light circulates in the sphere by WGM, but in this result, the anomalous Raman spectra is completely disappeared.

For further analysis of this observation, we performed a two-dimensional (2D) Raman spectrum scanning of the image in Fig. 1c, and Raman images were calculated using the Raman peaks of Si ( $525\text{ cm}^{-1}$ ), PS ( $1,006\text{ cm}^{-1}$ ) and the anomalous Raman spectrum ( $3,438\text{ cm}^{-1}$ ) (Fig. 1g–i). In the Si peak image of Fig. 1g, the Si frame on the right side showed high-intensity region. At the edge of the Si frame of the image centre, the Si spectrum was enhanced, which is an effect by conventional SERS<sup>13,19</sup>. At the PS peak of  $1,006\text{ cm}^{-1}$ , as expected, all the spheres in the scanned area showed a spherical shape similar to the optical microscopy (OM) images (Fig. 1h). Interestingly, with the peak of the anomalous Raman spectrum at  $3,438\text{ cm}^{-1}$ , the ring-like shapes were observed only for the spheres on the SiN film without direct contact with the Si frame, but not at all for those with contact (Fig. 1i). Furthermore, anomalous Raman spectra

were not observed for spheres on glass (Supplementary Fig. 1).

Next, we analysed the Raman images of the spheres on the SiN film without direct contact with the Si frame (Figs 2 and Supplementary Fig. 2). The Raman spectrum of the sphere periphery on the SiN film showed a very large signal with periodic intensity changes (Supplementary Fig. 2a–c). The top-view 2D Raman images of the microsphere showed a dome-shaped PS peak at  $1,006\text{ cm}^{-1}$  and a ring-shaped anomalous Raman spectrum peak (Supplementary Fig. 2d top). From the side, the Raman images of the PS peak and reflecting peak of  $72\text{ cm}^{-1}$  showed a spherical shape similar to the top view image, and the anomalous Raman image was clearest at the equatorial point of the spheres (Supplementary Fig. 2d bottom). For the spheres on the SiN film with Si frame contact, the anomalous Raman spectrum was hardly visible and no ring structure was observed (Supplementary Fig. 3). In the superimposed image of the PS peak and the anomalous Raman peak, the anomalous Raman spectrum was found to cover the surface of the sphere (Fig. 2a and b). Along a line drawn through the sphere, the PS signal of  $1,006\text{ cm}^{-1}$  showed a dome-like shape, while the anomalous Raman spectrum peak of  $3,438\text{ cm}^{-1}$  showed sharp peaks on both sides of the sphere (Fig. 2c). The width at half maximum of the PS signal was  $3.3\text{ }\mu\text{m}$ , almost equal to the sphere diameter of  $3\text{ }\mu\text{m}$ . The width between the two anomalous Raman spectrum peaks was

slightly larger, 3.5  $\mu\text{m}$ , indicating that this Raman spectrum existed only on the surface of the spheres. This result indicates that the incident light covers the circumference of the PS sphere. Therefore, the incident light is considered to be orbiting outside the perimeter of the sphere. Furthermore, the periodicity and phase of the anomalous Raman spectra differed from sphere to sphere (Fig. 2d). These differences may be due to the diameter of the spheres.

Based on these experimental results, we propose that the anomalous Raman spectrum was generated by irradiated light orbiting around the surface of dielectric spheres (Fig. 2e). When light is incident on a sphere, a part of the waveform on one side of the photon enters the sphere and bends its path along the curved surface (Fig. 2e). The refractive index of the PS sphere is approximately 1.6<sup>28,29</sup>, higher than that of air; thus, the light bends along the surface of the sphere upon reaching the surface. The light orbiting around the sphere surface travels towards the SiN film at the bottom, where it should continue its orbit without being scattered and then returns to the top (Fig. 2f). In addition, the wavelength of the circumferential light is expected to gradually shift to longer wavelengths upon travelling around the dielectric sphere due to gradual loss of energy. For spheres on glass or SiN film with Si frame contact, the orbiting light is scattered by the glass or Si frame, but does not return to the top of the sphere (Fig. 2g).

When two or three spheres on a SiN film are in close proximity, the anomalous Raman spectra show attenuation areas (Supplementary Fig. 4a–j). These attenuation areas are located on the line connecting the sphere centres and the sphere contact position (Supplementary Fig. 4d and i). This is because the incident light circulates around the surface of the spheres and is attenuated at the contact position of the spheres on the opposite side (Supplementary Fig. 4k). This result also strongly supports the phenomenon of light circling around the spheres.

#### *SiN thickness dependence of anomalous Raman spectrum*

Light orbiting on the sphere can continue its orbit without being scattered on the SiN film. To verify this mechanism, we measured and compared anomalous Raman spectra of spheres on SiN films of different thickness (Fig. 3). If the light orbiting the surface of the sphere simply transmits through the SiN film, the intensity of light transmission will increase with a thinner SiN film. Therefore, the intensity of the anomalous Raman spectrum should increase when the thinner SiN film, and decrease with increasing SiN thickness. Fig. 3a and b show the Raman spectra at the centre and periphery of 3  $\mu\text{m}$  spheres on SiN films of 20 to 100 nm thickness. The anomalous Raman spectrum around the sphere was the largest for the 50 nm thickness, and was

hardly detected for the SiN films of 20 or 100 nm thickness (Fig. 3a). At the sphere centre, only the Raman spectra of PS signal were seen for any film thickness (Fig. 3b). In the 3D pseudo-colour map of the spheres, the anomalous Raman spectrum was clearly seen surrounding the sphere surface on 50 nm thick SiN films (Fig. 3c). However, the anomalous Raman spectrum decreased upon varying the thickness from 50 nm (Fig. 3c and d).

These results suggest that the circumferential light does not simply pass through the SiN film but that there must be a mechanism that assists the light circulating most efficiently at 50 nm thickness. For such an assisted mechanism, we consider that the circulating light around the sphere enters the SiN film and returns to the sphere periphery by total reflection at the SiN bottom surface (Fig. 3e–g). When the SiN film is 20 nm thick, the light entering the SiN film continues to travel straight through without total reflection at the bottom surface (Fig. 3e). When a SiN film is 50 nm thick, the light entering the SiN film is totally reflected on the bottom surface and returns to the close neighbour of the sphere attachment point (Fig. 3f). This way, the light returns to the sphere surface and continues to circulate. When a SiN film is 100 nm thick, the photon entering the SiN film is totally reflected and returns to a place far from the sphere attachment point (Fig. 3g). The proposed mechanism explains why the anomalous

Raman spectrum is the most prominent at a SiN thickness of 50 nm.

In order to investigate the total reflection of the light at the bottom of the 50 nm thick SiN film, the Raman spectra of the spheres were measured with either water or immersion oil underneath the SiN film in place of air (Fig. 4). The pure water and immersion oil used here transmit light well, but their refractive indices of 1.33 and 1.52 are higher than that of air at 1.0. Therefore, the total reflection condition of light on the lower surface of the Si film is harder than that of air, and the anomalous Raman spectrum is expected to be reduced. The Raman spectral intensity around the sphere was significantly attenuated when water was filled underneath the SiN film (Fig. 4a–e). The spectral intensity was regained when the water was removed and replaced with air. When oil was filled underneath the SiN film, the Raman spectra around the spheres disappeared completely (Fig. 4f–h). This result indicated that the light continued to circulate due to the total reflection at the bottom surface of the SiN film. Furthermore, the lower surface of the SiN film under the sphere is extremely sensitive and functions as a high-sensitivity sensor because the circulating light is concentrated and totally reflected.

The refractive index of the SiN is approximately 2.0<sup>30,31</sup>, and at the interface with the air, the angle of total reflection is 30°, so that light entering from the periphery of the

sphere can easily be totally reflected (Supplementary Fig. 5a). However, if water or immersion oil is present underneath the SiN film, the refractive indices are 1.33 and 1.52, respectively; thus, the scattering angle of total reflection increases to  $41.7^\circ$  and  $49.4^\circ$ , and the light travels through without total reflection (Supplementary Fig. 5b and c). From these results and consideration, the angle of incident light under the SiN film is expected to be  $30^\circ$ – $41.7^\circ$ .

#### *Sphere diameter and anomalous Raman spectrum*

By examining the sphere diameter with the maximum intensity of the anomalous Raman spectrum, it is possible to compare the conditions for the generation of the circumferential light with the mathematical model. In addition, by investigating the cycle of the anomalous Raman spectrum intensity and the sphere diameter, the generation mechanism can be determined. We obtained the Raman spectra of spheres with diameters of 2–7  $\mu\text{m}$  on a 50 nm thick SiN film (Fig. 5a). With a sphere diameter of 3 and 4  $\mu\text{m}$ , the anomalous Raman spectrum around the sphere was significantly increased. (Fig. 5a and b). In contrast, with a sphere diameter of 2  $\mu\text{m}$  and 5  $\mu\text{m}$  or more, the anomalous Raman spectrum was very weak. In the 3D pseudo-colour map of the spheres, the anomalous Raman spectrum was clearly seen surrounding the sphere with

the diameter of 3 and 4  $\mu\text{m}$  (Fig. 5c). For other diameters, the spectra were very weak. Fig. 5d shows the normalised Raman spectrum of the sphere periphery after subtracting the PS Raman spectrum at the sphere centre. For a diameter of 2  $\mu\text{m}$ , the Raman spectrum changed with a slow cycle, approximately 4 cycles (Fig. 5d top). The periodicity of this spectrum gradually increased with an increasing sphere diameter, reaching 11 cycles with a diameter of 7  $\mu\text{m}$ . These gentle and periodic changes of the Raman spectrum are presumably due to interference<sup>32</sup>. The orbiting photon around the spheres is expected to be interfered with in the process of repeated orbiting on the sphere surface (Fig. 5e). Model calculations show that the periodicity of this interference increases with increasing diameter of the sphere (Fig. 5f).

The relationship between optical power intensity  $P(\lambda)$  and interference is expressed by the following equation (see details under Materials and Methods).

$$P(\lambda) = R(\lambda) \left[ \frac{1}{n} \sum_{m=1}^n \sin \left( 2\pi \frac{\pi m D_s \eta_s}{\lambda} \right) + 1 \right]^2 \quad (1)$$

Here,  $\lambda$  is wavelength,  $R(\lambda)$  is detection efficiency relative to wavelength,  $D_s$  is the sphere diameter,  $\eta_s$  is refractive index of PS of 1.6<sup>28,29</sup>, and  $n$  is the number of light orbits and is set to 2. Fig. 5f shows the optical power of orbiting light calculated by

equation (1) at wavelengths between 532 and 640 nm and for sphere diameters between 2 and 7  $\mu\text{m}$ . For a diameter of 2  $\mu\text{m}$ , the change in light intensity is about 4 cycles in the numerical model (Fig. 5f top). In the case of diameters of 3 to 7  $\mu\text{m}$ , the periodicity of the intensity change was 5 to 11 (Fig. 5f), which agreed with the experimental results (Fig. 5d). However, the waveform on the positive side of the model rises slightly more sharply than the experimental results. Because the spheres used in the experiments were not perfectly spherical, a slight difference in length can be expected depending on the direction of the circumferential light, resulting in a broad interference waveform.

#### *Comparison of fluorescence images of spheres attached to the SiN film and glass*

Finally, we compared the fluorescence images of PS spheres attached to the SiN film and glass using a fluorescence microscope (Fig. 6). The Raman spectrum around the spheres showed a gradual shift to longer wavelengths than the incident laser at 532 nm (Figs 1 and 2). Therefore, it is expected that the circumferential light that shifts to longer wavelengths than the excitation light can be observed using a conventional fluorescence microscope. The 3  $\mu\text{m}$  PS spheres attached to the 50-nm thick SiN film were viewed under a fluorescence microscope, and the clear fluorescence images were observed with excitation light between 365 and 565 nm (Fig. 6a–d). In contrast, the

spheres attached to the slide glass showed weak blue spherical autofluorescence with excitation light of 365 nm only (Fig. 6e and f). With excitation light of longer wavelengths such as 480 and 565 nm, the spherical shape completely disappeared (Fig. 6g and h). Since the spheres used in this experiment did not contain any fluorescent dye, only weak autofluorescence should normally be observed e.g. in the case of spheres attached to glass (Fig. 6f). Therefore, the fluorescence image of spheres on the SiN film certainly reflected light circulating around the sphere.

In the enlarged image of a single sphere on the SiN film, the ring-shaped fluorescence images were observed at any excitation wavelength (Fig. 6i). In contrast, in the case of the glass substrate, a weak spherical fluorescence image was observed only with 365 nm excitation light (Fig. 6j). Along the line going through the centre of the sphere at the SiN film with an excitation wavelength of 365, sharp peaks were seen at the boundary surfaces at both ends of the sphere at each detected intensity of red, green, and blue (Fig. 6k left). On the glass substrate, a dome-like shape showed the blue line plot, and green and red line plots showed just noise (Fig. 6k right). These results indicate that the excitation incident light on the spheres was gradually shifted to longer wavelengths while circulating around the outside surface of the sphere.

## Discussion

The photons orbiting around the surface of the dielectric microspheres according to the proposal presented here have different characteristics from the WGMs orbiting inside the spheres. WGM produces many sharp light intensity peaks and/or Raman spectrum peaks due to the resonance effect caused by the light circulating inside the fluorescence-doped spheres<sup>8,21,23</sup>. In our study, anomalous periodic Raman spectra were observed from PS microspheres without dye on SiN films, caused by interference of light orbiting outside the spheres. (Figs 1 and 5). In general, light interference is produced by light reflected from interfaces and thin films; this has applications in interferometers and Fabry-Perot etalons. However, the interference phenomenon caused by light orbiting on the sphere surface, which has been found for the first time in this study, should have a wide range of applications for micro-optical devices, e.g. micro interferometers, small ultra-sensitive sensors, optical modulators and nonlinear optical qubits.

The conditions for light to orbit on the sphere surface are determined by the refractive index and the diameter of the sphere (equation (2)).

$$D_s' = d \frac{\eta_s + \eta_a}{\eta_s - \eta_a} \quad (2)$$

Here  $D_s'$  is the theoretical diameter of the sphere that satisfies the circumference condition,  $d$  is photon waveform width (Fig. 7), and  $\eta_s$  and  $\eta_a$  are refractive indices of the PS sphere, approximately 1.6, and that of air, 1.0, respectively (see details under Materials and Methods). If we set  $d$  to 530 nm, the diameter  $D_s'$  is 2.17  $\mu\text{m}$ , which is smaller than the experimental result where the circumference light intensity is highest when the diameter  $D_s'$  is 3  $\mu\text{m}$  (Fig. 5). This difference is explained by the actual refractive index of the PS microsphere being smaller than the bulk PS. From the experimental results, the refractive index of the PS microsphere would be expected to be 1.43.

There are two possible explanations for the shift to longer wavelengths of the excitation light orbiting around a sphere: one is a nonlinear effect whereby only one side of the photon waveform is trapped on the surface of a dielectric sphere and travels around it, and the other is energy loss due to orbiting around a microsphere at light speed. In order to distinguish between these explanations, it will be necessary to evaluate the change in terms of the duration and wavelength of the orbiting photon using a femtosecond pulsed laser. In addition, theoretical investigations based on quantum electromagnetism will be necessary.

The light orbiting around dielectric microspheres may affect the visibility of clouds in the sky. Clouds are composed of small water and/or ice droplets, which can be regarded as dielectric microspheres. Therefore, light is expected to orbit around the circumference of the water droplets. The short wavelength light, including ultraviolet light, increases its wavelength upon orbiting around water droplets. Accordingly, clouds at high altitudes that receive strong ultraviolet irradiation may be more clearly visible due to the augmentation of light intensity from the circumferential light and the Mie-scattering phenomenon.

In conclusion, we report for the first time that light orbits around the surface of dielectric spheres. This circumferential light was observed most strongly when dielectric spheres of 3  $\mu\text{m}$  diameter were placed on a 50-nm thick SiN film. The excitation light incident to the surface of PS spheres gradually shifts to a longer wavelength while orbiting outside of the sphere surface. Furthermore, interference among the circumferential lights produces a gradual periodic change in the Raman spectrum. These physical properties can be applied to high-sensitivity sensors, modulators, and quantum devices. Finally, our study uses ordinary materials such as PS spheres and SiN films as well as conventional measurement systems. In the near future, the system described here is expected to be widely utilized as a general-purpose

platform for new optical devices.

## **Materials and Methods**

### **Sample preparation**

The PS microspheres of 2–7  $\mu\text{m}$  diameter suspended in aqueous buffer (Micromer, micromod Partikeltechnologie GmbH, Germany) were diluted three times with ultrapure water, vortexed for 10 s and sonicated for 1 min. The microsphere suspension (5  $\mu\text{L}$ ) was placed on a SiN film supported by a Si frame attached on the aluminium holder<sup>33</sup>, and 10 s later, the suspension liquid was absorbed by a piece of filter paper from the droplet. On a glass substrate, 10  $\mu\text{L}$  of the microsphere suspension was placed on a slide glass (Micro slide glass S1111, Matunami Glass Ind. Ltd., Japan), and after absorbing the suspension liquid from the droplet by filter paper, the sample was dried for 5 min at a room temperature of 23 °C.

The Raman spectra of the PS microspheres were measured with air and water or oil underneath the SiN film. In the measurement condition of water or oil on the underneath the Si film, 5  $\mu\text{L}$  ultrapure water or immersion oil (Type HF code 368, Refractive index 1.51–1.52, Cargille Labs. Inc., USA) was dropped into the centre hole of the aluminium

holder, filled the underneath the Si film, and then the hole was covered with Kapton tape (tesa 51408, Tesa tape Inc., USA).

### **Raman microscopy**

The PS microspheres on a SiN film or slide glass were observed under a confocal Raman microscope using a  $\times 100$  objective lens (Plan apo, Carl Zeiss, Oberkochen, Germany) and a 532-nm Nd-YAG laser (alpha300R, WITech, Ulm, Germany). Spectra were acquired with a Peltier-cooled charge-coupled device detector (DV401-BV, Andor, UK) with 600 gratings/mm (UHTS 300VIS, WITec, Germany). WITec suite (version 7.0, WITec, Germany) was used for data acquisition. For 2D Raman spectra, the laser intensity was 1.5–3 mW and the number of pixels in the XY axis was from 120 $\times$ 120 to 300 $\times$ 300. The pixel step width was 100 nm, and the measurement time for each pixel was set to 0.05 s. XZ axis scans were performed from 120 $\times$ 60 pixels, the step width set to X axis of 100 nm and Z axis of 200 nm. Raman spectral data were calculated using MATLAB R2020a (Math Works Inc., Natick, MA, USA) and plotted using Origin 2021J (Origin-Lab Co., Northampton, MA, USA).

### **Calculation of Raman spectrum and 2D Raman image of PS spheres**

The 2D Raman spectral data of the spheres were converted to the Matlab data file using WITec suite and were transferred to a personal computer (Intel Core i7, 3.2 GHz, Windows 10). The 2D Raman spectral images were calculated by Matlab R2020a with Image Processing Toolbox and Signal Processing Toolbox. The average Raman spectrum of the sphere centre was calculated from the pixels within a circle with a radius of 5 pixels from the sphere centre. For the spectrum of the sphere periphery, the Raman image of the PS peak of  $1,006\text{ cm}^{-1}$  was normalized after applying a  $0.5\sigma$  Gaussian filter, and Raman spectra of the sphere periphery were averaged from pixels with normalized PS peak intensities between 0.2 and 0.5.

### **Optical phase microscopy and fluorescence imaging.**

The microspheres of  $3\text{ }\mu\text{m}$  in diameter attached to a slide glass (Matunami, Japan) and a SiN film were visualised at  $\times 400$  magnification using an optical phase contrast microscope (AXIO Observer A1; Carl Zeiss, Oberkochen, Germany). Fluorescence images of the spheres were observed using fluorescence filters of the excitation/emission wavelength at  $365/420\text{ nm}$ ,  $480/520\text{ nm}$  and  $565/620\text{ nm}$  (Filter Set 02, 04 and 31, respectively, Carl Zeiss, Germany). The observed fluorescence images were normalised to an 8-bit scale by the maximum and minimum brightness intensity in

each image. However, observation of spheres attached to glass slides using excitation light of 480 nm and 565 nm resulted in the disappearance of the sphere image. Therefore, the original images were shown in Ex480 and Ex565 of Fig. 6g, h and j to avoid noise enhancement due to normalization.

### **Calculation of interference by orbiting light**

The circumference of a sphere with diameter  $D_s$  is  $\pi D_s$ . Interference due to light circulating around the spheres is assumed to be produced by light that has travelled around the sphere once and twice ( $n=2$ ). With the wavelength and sphere reflectance being  $\lambda$  and  $\eta_s$ , the light intensity  $I(\lambda)$  is given by the following equation.

$$I(\lambda) = \frac{1}{n} \sum_{m=1}^n \sin\left(2\pi \frac{\pi m D_s \eta_s}{\lambda}\right) + 1 \quad (3)$$

The range of this equation is  $\pm 1$ , which corresponds to the normalized light intensity. Since the light intensity does not take a negative value, we added 1 to make it greater than 0, and squared it to produce the light power (equation (1)). In the Raman spectrum of the PS sphere periphery, the anomalously enhanced Raman intensity is weak at short wavelengths, and the amplitude of the spectrum increases gradually with wavelength

getting longer (Fig. 5). This wavelength-dependent optical enhancement characteristic is denoted by  $R(\lambda)$  and is approximated by an exponential saturation characteristic.

$$R(\lambda) = 1 - \exp\left(-\frac{\lambda - \lambda_i}{\tau}\right) \quad (4)$$

Here  $\lambda_i$  is the wavelength of the incident laser, 532 nm, and  $\tau$  is the time constant of the wavelength ( $\tau=100$ ). The calculation of  $I(\lambda)$ ,  $R(\lambda)$  and  $P(\lambda)$  for a 3  $\mu\text{m}$  sphere with a wavelength between 532 and 640 nm is shown in Supplementary Fig. 6. The numerical calculations were performed using a PC (Windows10, 3.2 GHz, 8 core CPU: Core-i7) with Matlab R2020a.

### **Circumferential light conditions for dielectric spheres**

The amplitude width of the incident photon is  $d$ , and it is assumed that half of it travels inside the sphere (Fig. 7). Under such an assumption, the speed of light travelling inside the PS sphere is decelerated by  $1/\eta_s$  ( $\eta_s=1.6$ ), and in the air layer, the light speed is normal ( $\eta_a=1$ ). The circumference of light around the dielectric sphere arises when the difference between the length of the circumference inside the sphere and the outside the sphere is balanced by the light speed inside and outside. This can be formulated as

follows.

$$2\pi \left( \frac{D_s}{2} - \frac{d}{2} \right) \eta_s = 2\pi \left( \frac{D_s}{2} + \frac{d}{2} \right) \eta_a \quad (5)$$

From the above equation (5), the sphere diameter  $D_s$ , is calculated using equation (2).

### **Reporting summary.**

Further information on research design is available in the Nature Research Reporting Summary linked this article.

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### **Data availability**

All data generated or analysed during this study are presented in this paper or in the Supplementary Information. All the raw data files or spectra are available from the corresponding authors on reasonable request.

### **Author Contributions**

T. O. designed and performed the research. T. O. contributed to obtaining the experimental data and writing the manuscript. All authors read and approved the manuscript.

### **Competing interests**

The authors have declared that no competing of interest exists.

### **Additional information**

Supplementary information is available for this paper at <http://doi.org/XX.XXX/XXXX>.

### **References**

1. Vollmer, F. et al. Protein detection by optical shift of a resonant microcavity.

- Appl. Phys. Lett.* **80**, 4057–4059 (2002).
2. Armani, A. M., Kulkarni, R. P., Fraser, S. E., Flagan, R. C. & Vahala, K. J. Label-free, single-molecule detection with optical microcavities. *Science* **317**, 783–787 (2007).
  3. Baaske, M. D. & Vollmer, F. Optical observation of single atomic ions interacting with plasmonic nanorods in aqueous solution. *Nat. Photon.* **10**, 733–739 (2016).
  4. Heshmati, S., Abedi, K. & Darvish, G. High-Q microsphere integrated with a tapered fiber suitable for biosensing applications. *Opt. Quant. Elect.* **51**, 273 (2019).
  5. Gorodetsky, M. L. & Ilchenko, V. S. Optical microsphere resonators: optimal coupling to high-Q whispering-gallery modes. *J. Opt. Soc. Am. B* **16**, 147–154 (1999).
  6. Ilchenko, V. S., Yao, X. S. & Maleki, L. Pigtailling the high-Q microsphere cavity: a simple fiber coupler for optical whispering-gallery modes. *Opt. Lett.* **24**, 723–725 (1999).
  7. Spillane, S. M., Kippenberg, T. J., Painter, O. J. & Vahala, K. J. Ideality in a fiber-taper-coupled microresonator system for application to cavity quantum

- electrodynamics. *Phys. Rev. Lett.* **91**, 043902 (2003).
8. Ghulinyan, M. et al. Whispering-gallery modes and light emission from a Si-nanocrystal-based single microdisk resonator. *Opt. Express* **16**, 13218–13224 (2008).
  9. Zhao, Q., Zhou, J., Zhang, F. L. & Lippens, D. Mie resonance-based dielectric metamaterials. *Mater. Today* **12**, 60–69 (2009).
  10. Fu, Y. H., Kuznetsov, A. I., Miroshnichenko, A. E., Yu, Y. F. & Luk'yanchuk, B. Directional visible light scattering by silicon nanoparticles. *Nat. Commun.* **4**, 1527 (2013).
  11. Preston, T. C. & Reid, J. P. Determining the size and refractive index of microspheres using the mode assignments from Mie resonances. *J. Opt. Soc. Am. A* **32**, 2210–2217 (2015).
  12. Savo, R. et al. Broadband Mie driven random quasi-phase-matching. *Nat. Photon.* **14**, 740–747 (2020).
  13. Nie, S. M. & Emery, S. R. Probing single molecules and single nanoparticles by surface-enhanced Raman scattering. *Science* **275**, 1102–1106 (1997).
  14. Haynes, C. L. & Van Duyne, R. P. Plasmon-sampled surface-enhanced Raman excitation spectroscopy. *J. Phys. Chem. B* **107**, 7426–7433 (2003).

15. Deng, Z., Chen, M. & Wu, L. Novel method to fabricate SiO<sub>2</sub>/Ag composite spheres and their catalytic, surface-enhanced Raman scattering properties. *J. Phys. Chem. C* **111**, 11692–11698 (2007).
16. Alessandri, I. Enhancing Raman scattering without plasmons: Unprecedented sensitivity achieved by TiO<sub>2</sub> shell-based resonators. *J. Am. Chem. Soc.* **135**, 5541–5544 (2013).
17. Kuznetsov, A. I., Miroschnichenko, A. E., Brongersma, M. L., Kivshar, Y. S. & Luk'yanchuk, B. Optically resonant dielectric nanostructures. *Science* **354**, aag2472 (2016).
18. Alessandri, I. & Lombardi, J. R. Enhanced Raman scattering with dielectrics. *Chem. Rev.* **116**, 14921–14981 (2016).
19. Raza, S. & Kristensen, A. Raman scattering in high-refractive-index nanostructures. *Nanophotonics* **10**, 1197–1209 (2021).
20. Baer, T. Continuous-wave laser oscillation in a Nd:YAG sphere. *Opt. Lett.* **12**, 392–394 (1987).
21. Kuwata-Gonokami, M., Takeda, K., Yasuda, H. & Ema, K. Laser-emission from dye-doped polystyrene microsphere. *Jpn. J. Appl. Phys.* **31**, L99–L101 (1992).
22. McCall, S. L., Levi, A. F. J., Slusher, R. E., Pearton, S. J. & Logan, R. A.

- Whispering-gallery mode microdisk lasers. *Appl. Phys. Lett.* **60**, 289–291 (1992).
23. Sandoghdar, V. et al. Very low threshold whispering-gallery-mode microsphere laser. *Phys. Rev. A* **54**, R1777–R1780 (1996).
24. Spillane, S. M., Kippenberg, T. J. & Vahala, K. J. Ultralow-threshold Raman laser using a spherical dielectric microcavity. *Nature* **415**, 621–623 (2002).
25. Campillo, A. J., Eversole, J. D. & Lin, H. B. Cavity quantum electrodynamic enhancement of stimulated emission in microdroplets. *Phys. Rev. Lett.* **67**, 437–440 (1991).
26. Vernooy, D. W., Furusawa, A., Georgiades, N. P., Ilchenko, V. S. & Kimble, H. J. Cavity QED with high-Q whispering gallery modes. *Phys. Rev. A* **57**, R2293–R2296 (1998).
27. Armani, D. K., Kippenberg, T. J., Spillane, S. M. & Vahala, K. J. Ultra-high-Q toroid microcavity on a chip. *Nature* **421**, 925–928 (2003).
28. Nikolov, I. D. & Ivanov, C. D. Optical plastic refractive measurements in the visible and the near-infrared regions. *Appl. Optics* **39**, 2067–2070 (2000).
29. Naglic, P., Zielinsky, Y., Likar, B. & Burmen, M. Determination of refractive index, size, and solid content of monodisperse polystyrene microsphere

- suspensions for the characterization of optical phantoms. *Biomed. Opt. Express* **11**, 1901–1918 (2020).
30. Kessels, W. M. M. et al. High-rate deposition of a-SiN<sub>x</sub> : H for photovoltaic applications by the expanding thermal plasma. *J. Vac. Sci. Technol. A* **20**, 1704–1715 (2002).
31. Verlaan, V. et al. The effect of composition on the bond structure and refractive index of silicon nitride deposited by HWCVD and PECVD. *Thin Solid Films* **517**, 3499–3502 (2009).
32. Stavenga, D. G. Thin film and multilayer optics cause structural colors of many insects and birds. *Mater. Today* **1**, 109–121 (2014).
33. Ogura, T. Nanoscale analysis of unstained biological specimens in water without radiation damage using high-resolution frequency transmission electric-field system based on FE-SEM. *Biochem. Biophys Res. Commun.* **459**, 521–528 (2015).

## Figure Legends

**Fig. 1 Raman spectrum of PS microspheres on SiN film. a** Schematic of the setup

used to measure the Raman spectrum of 3  $\mu\text{m}$  diameter PS spheres on a SiN film of 50 nm thickness supported by a Si frame. **b** OM image of the spheres on a  $0.4 \times 0.4$  mm square SiN film. **c** Higher magnification OM image ( $\times 1,000$ ) of the boundary region between the SiN film and the Si frame (dashed red rectangle in **b**). **d** Raman spectra of the sphere centre (red line) and left periphery (blue line) on the SiN film indicated by a red arrow in **c**. The sphere centre exhibited a PS spectrum, while the periphery showed a very high intensity spectrum with periodicity. **e** Raman spectrum of **d** focusing on a range of  $800\text{--}1,200$   $\text{cm}^{-1}$ . At the sphere centre, the PS Raman peak of  $1,006$   $\text{cm}^{-1}$  was clear (red line), while the PS peak of the sphere periphery was very low (blue line). **f** Raman spectra of the sphere centre (red line) and left periphery (blue line) on the SiN film contacted by a Si frame indicated by a blue arrow in **c**. Under this condition, similar PS and Si peaks were observed in both Raman spectra. **g** Raman image of Si peak ( $525$   $\text{cm}^{-1}$ ) in **c**. The Si frame on the right side was clearly observed. **h** With the Raman image of PS peak ( $1,006$   $\text{cm}^{-1}$ ), all spheres in the scanned area were clearly observed. **i** With the Raman image of  $3,438$   $\text{cm}^{-1}$ , the spheres on the SiN film showed a ring shape. Scale bars,  $10$   $\mu\text{m}$  in **c** and **g**.

**Fig. 2 Raman spectra of the sphere circumference on SiN film. a, b** Top and side

views of merged Raman images of the PS peak ( $1,006\text{ cm}^{-1}$ , red) and anomalous peak of  $3,438\text{ cm}^{-1}$  (green) with a  $3\text{ }\mu\text{m}$  diameter sphere on a  $50\text{ nm}$  thick SiN film without contact with a Si frame. The PS sphere is seen to be covered with a ring-shaped anomalous Raman peak. **c** Comparison of the line plots of PS peak ( $1,006\text{ cm}^{-1}$ , red) and  $3,438\text{ cm}^{-1}$  (green) at the sphere centre in **a**. The Raman intensities are normalized individually. The distance between the two half maxima of the PS peak is  $3.3\text{ }\mu\text{m}$  and the distance between the peaks at the periphery of the  $3,438\text{ cm}^{-1}$  is  $3.5\text{ }\mu\text{m}$ . **d** Comparison of the Raman spectrum at two sphere circumferences in Fig. 1*i* indicated by red and blue arrows. Both spectra were of 4 cycles but the phases were different. **e** Schematic of the generation mechanism of the periodic high intensity Raman spectrum at the periphery of the dielectric sphere in air. When light is incident on the sphere surface, one side of the photon waveform enters the dielectric sphere and orbits the outside surface. **f** In the spheres on a SiN film, the light continues to circulate without being affected by the film. **g** In the spheres on a SiN film directly on a Si frame, the orbiting light is scattered and lost due to the influence of the Si frame. Scale bar,  $3\text{ }\mu\text{m}$  in **a**.

**Fig. 3 Relationship between SiN thickness and the intensity of the sphere's**

**circumferential light.** **a** The average Raman spectra of the circumference of the 3  $\mu\text{m}$  diameter PS spheres with a SiN thickness of 20–100 nm. The circumferential light was most intense with the SiN film of 50 nm thickness but hardly detectable with a SiN film of 20 or 100 nm thickness. **b** Average Raman spectrum at the sphere centre. The same PS spectra were seen for all the SiN film thicknesses. **c** Pseudo-3D colour maps of the PS peaks and the anomalous Raman peaks for SiN thicknesses of 20–100 nm. The maps of the PS peak showed a similar dome-like shape for all SiN thicknesses (bottom row). With the anomalous Raman peaks, the ring shape around the spheres was clearly observed at the SiN thicknesses of 50 and 75 nm (top row). **d** Relationship between the anomalous Raman peaks and SiN thickness. Six spheres were measured at each SiN thickness and the ratios of the maximum anomalous Raman intensity to that of the PS peak ( $1,006\text{ cm}^{-1}$ ) at the sphere centre were calculated and normalised. Anomalous Raman intensity had a maximum at 50 nm SiN thickness but was reduced when the SiN thickness was increased or decreased. **e** Conceptual diagram of dependence of the circumferential light intensity of a dielectric microsphere on the SiN thickness. With a 20 nm thick SiN film, the light travelling downwards around the periphery of the sphere enters the SiN film and exits downwards. **f** With a 50 nm thick SiN film, the light entering the SiN film from a sphere is totally reflected at the bottom surface of the SiN

film and returns to a point next to the sphere attachment point. The light returns to the sphere surface and continues to circulate. **g** With a 100 nm thick SiN film, the light that enters the SiN film and is totally reflected returns to a point far from the sphere attachment point. Scale bar, 3  $\mu\text{m}$  in **c**.

**Fig. 4 Raman spectrum of PS microspheres on the SiN film with water or oil underneath the film instead of air.**

**a** An OM image of 3 $\mu\text{m}$  diameter PS spheres on the 50 nm thick SiN film. **b** Magnified sphere image indicated by a red arrow in **a**. **c** Schematic of the measurement of Raman spectrum of 3  $\mu\text{m}$  diameter PS spheres on a SiN film with water or immersion oil underneath the film. **d** Pseudo-3D colour maps of the anomalous Raman peaks on a SiN film with water or oil underneath the film. **e** Pseudo-3D colour maps of PS Raman peaks on a SiN film with water or oil underneath of the film. The sphere structures were observed in all the cases. **f** Raman spectra of sphere peripheries normalized by PS peaks of 1,006  $\text{cm}^{-1}$  of the sphere centre on a SiN film with air, water or oil underneath the film. The anomalous periodic Raman spectra are seen with the air underneath the film (red, with air in **c** and **d** left and blue, with removal of water in **c** and **d** the third from the left). With water underneath the film, the anomalous Raman spectra decreased

(purple, with water, in **c** and **d** the second from the left). With oil underneath the film, the Raman spectra indicated only the PS spectrum (green, with oil, in **c** and **d** right). **g** Normalized Raman spectra of sphere centre by PS peaks of  $1,006\text{ cm}^{-1}$  at the sphere centre on a SiN film with air, water or oil underneath the film. The Raman spectra under any condition show only the PS spectrum. **h** Relationship between the anomalous Raman peaks around the spheres and the SiN film with air, water or oil underneath the film. Six sphere samples were measured under each condition. Scale bars,  $10\text{ }\mu\text{m}$  in **a**,  $3\text{ }\mu\text{m}$  in **b** and **d**.

**Fig. 5 Sphere size and circulating light intensity.** **a** Averaged Raman spectra of the circumference of PS spheres of  $2\text{--}7\text{ }\mu\text{m}$  diameter on a  $50\text{ nm}$  thick SiN film. **b** Relationship between the anomalous Raman peaks and sphere diameter. Six spheres were measured at each diameter and the ratio of the maximal anomalous Raman intensity to the PS peak ( $1,006\text{ cm}^{-1}$ ) at the sphere centre was calculated and normalised. The anomalous Raman peak intensity was the highest at the sphere diameter of  $3\text{ }\mu\text{m}$  but was very low at  $2\text{ }\mu\text{m}$  and  $5\text{--}7\text{ }\mu\text{m}$ . **c** Pseudo-3D colour maps of the PS peak and the circumferential peak for sphere diameter of  $2\text{--}7\text{ }\mu\text{m}$ . The maps of the PS peak show similar dome-like shape for all diameters (bottom row). With the anomalous Raman

peak, the ring shape around the spheres was clearly observed at the SiN thickness of 3 and 4  $\mu\text{m}$  (top row). **d** Normalized Raman spectrum of the sphere circumference with a diameter of 2–7  $\mu\text{m}$ . The arrows in the figure indicate the peaks of each spectral periodicity. With the sphere diameter increment from 2 to 7  $\mu\text{m}$ , the periodicity of the Raman spectrum increases from 4 to 11. **e** Schematic of the interference produced by light circulating around the sphere. A photon travelling around the periphery of the sphere interferes with other photons while they travel around the sphere. **f** Raman spectrum of sphere periphery by the numerical model of equation (1). The sphere diameters of the model were set to the values where the period and phase were in close correspondence with the experimental results (left side diameters in each plot). Scale bar, 3  $\mu\text{m}$  in **c**.

**Fig. 6 Comparison of PS spheres attached to glass and SiN film by fluorescence microscopy.** **a** OM image of the 3  $\mu\text{m}$  diameter PS spheres on a 50 nm thick SiN film. **b–d** Images of spheres attached to the SiN film using three sets of fluorescence filters with excitation at 365 nm and fluorescence at 420 nm (Ex365/F420), excitation at 480 nm and fluorescence at 520 nm (Ex480/F520) and excitation at 565 nm and fluorescence at 620 nm (Ex565/F620). The PS spheres attached to the SiN film were

clearly observed with all three sets of filters. **e** OM image of the 3  $\mu\text{m}$  diameter spheres attached to the slide glass. **f–h** Images of spheres attached to the slide glass using three sets of fluorescence filters with excitation at Ex365/F420, Ex480/F520 and Ex565/F520. The spheres attached to the glass showed very weak autofluorescence with a set of filters of Ex365/F420, and the sphere structure was not discerned with set of filters of Ex480/F520 or Ex565/F620. **i** Magnified OM image and fluorescence filter images of spheres on the SiN film indicated by a white arrow in **b**. In all fluorescence filter images, ring-like structures were observed around the periphery of the spheres. **j** Magnified OM image and fluorescence filter images of the spheres attached to the slide glass indicated by a white arrow in **f**. **k** Comparison of the line plots at the centre of the sphere image using the set of filters, Ex365/F420, on the SiN film and glass. The red, blue and green lines correspond to the RGB intensity values at the white dotted line position at the sphere centre, respectively (dotted line in **i** and **j** Ex365). The normalised RGB intensities of PS sphere on a SiN film show sharp peaks at the sphere ends (left plot). On the other hand, for the spheres on the slide glass, the blue line shows a dome shape, but the red and green intensity show no spherical structure but just noisy (right plot). Scale bars, 10  $\mu\text{m}$  in **a** and 3  $\mu\text{m}$  in **j**.

**Fig. 7 Schematic of light circulating around a dielectric microsphere.**

The amplitude width of the incident photon to the dielectric sphere is  $d$ , and it is assumed that half of it extends into the inside of the sphere. If the diameter of the sphere is  $D_s$ , the innermost radius of the light circling the sphere is  $1/2(D_s - d)$ . On the other hand, the outermost radius of the light circling the sphere is  $1/2(D_s + d)$ .

# Figures

## Figure 1

Raman spectrum of PS microspheres on SiN film. a Schematic of the setup used to measure the Raman spectrum of 3  $\mu\text{m}$  diameter PS spheres on a SiN film of 50 nm thickness supported by a Si frame. b OM image of the spheres on a 0.4  $\times$  0.4 mm square SiN film. c Higher magnification OM image ( $\times 1,000$ ) of the boundary region between the SiN film and the Si frame (dashed red rectangle in b). d Raman spectra of the sphere centre (red line) and left periphery (blue line) on the SiN film indicated by a red arrow in c. The sphere centre exhibited a PS spectrum, while the periphery showed a very high intensity spectrum with periodicity. e Raman spectrum of d focusing on a range of 800-1,200  $\text{cm}^{-1}$ . At the sphere centre, the PS Raman peak of 1,006  $\text{cm}^{-1}$  was clear (red line), while the PS peak of the sphere periphery was very low (blue line). f Raman spectra of the sphere centre (red line) and left periphery (blue line) on the SiN film contacted by a Si frame indicated by a blue arrow in c. Under this condition, similar PS and Si peaks were observed in both Raman spectra. g Raman image of Si peak (525  $\text{cm}^{-1}$ ) in c. The Si frame on the right side was clearly observed. h With the Raman image of PS peak (1,006  $\text{cm}^{-1}$ ), all spheres in the scanned area were clearly observed. i With the Raman image of 3,438  $\text{cm}^{-1}$ , the spheres on the SiN film showed a ring shape. Scale bars, 10  $\mu\text{m}$  in c and g.

## Figure 2

Raman spectra of the sphere circumference on SiN film. a, b Top and side views of merged Raman images of the PS peak (1,006  $\text{cm}^{-1}$ , red) and anomalous peak of 3,438  $\text{cm}^{-1}$  (green) with a 3  $\mu\text{m}$  diameter sphere on a 50 nm thick SiN film without contact with a Si frame. The PS sphere is seen to be covered with a ring-shaped anomalous Raman peak. c Comparison of the line plots of PS peak (1,006  $\text{cm}^{-1}$ , red) and 3,438  $\text{cm}^{-1}$  (green) at the sphere centre in a. The Raman intensities are normalized individually. The distance between the two half maxima of the PS peak is 3.3  $\mu\text{m}$  and the distance between the peaks at the periphery of the 3,438  $\text{cm}^{-1}$  is 3.5  $\mu\text{m}$ . d Comparison of the Raman spectrum at two sphere circumferences in Fig. 1i indicated by red and blue arrows. Both spectra were of 4 cycles but the phases were different. e Schematic of the generation mechanism of the periodic high intensity Raman spectrum at the periphery of the dielectric sphere in air. When light is incident on the sphere surface, one side of the photon waveform enters the dielectric sphere and orbits the outside surface. f In the spheres on a SiN film, the light continues to circulate without being affected by the film. g In the spheres on a SiN film directly on a Si frame, the orbiting light is scattered and lost due to the influence of the Si frame. Scale bar, 3  $\mu\text{m}$  in a.

## Figure 3

Relationship between SiN thickness and the intensity of the sphere's circumferential light. a The average Raman spectra of the circumference of the 3  $\mu\text{m}$  diameter PS spheres with a SiN thickness of 20-100 nm. The circumferential light was most intense with the SiN film of 50 nm thickness but hardly detectable with a SiN film of 20 or 100 nm thickness. b Average Raman spectrum at the sphere centre. The same PS spectra were seen for all the SiN film thicknesses. c Pseudo-3D colour maps of the PS peaks and the anomalous Raman peaks for SiN thicknesses of 20-100 nm. The maps of the PS peak showed a similar dome-like shape for all SiN thicknesses (bottom row). With the anomalous Raman peaks, the ring shape around the spheres was clearly observed at the SiN thicknesses of 50 and 75 nm (top row). d Relationship between the anomalous Raman peaks and SiN thickness. Six spheres were measured at each SiN thickness and the ratios of the maximum anomalous Raman intensity to that of the PS peak ( $1,006\text{ cm}^{-1}$ ) at the sphere centre were calculated and normalised. Anomalous Raman intensity had a maximum at 50 nm SiN thickness but was reduced when the SiN thickness was increased or decreased. e Conceptual diagram of dependence of the circumferential light intensity of a dielectric microsphere on the SiN thickness. With a 20 nm thick SiN film, the light travelling downwards around the periphery of the sphere enters the SiN film and exits downwards. f With a 50 nm thick SiN film, the light entering the SiN film from a sphere is totally reflected at the bottom surface of the SiN film and returns to a point next to the sphere attachment point. The light returns to the sphere surface and continues to circulate. g With a 100 nm thick SiN film, the light that enters the SiN film and is totally reflected returns to a point far from the sphere attachment point. Scale bar, 3  $\mu\text{m}$  in c.

#### Figure 4

Raman spectrum of PS microspheres on the SiN film with water or oil underneath the film instead of air. a An OM image of 3 $\mu\text{m}$  diameter PS spheres on the 50 nm thick SiN film. b Magnified sphere image indicated by a red arrow in a. c Schematic of the measurement of Raman spectrum of 3  $\mu\text{m}$  diameter PS spheres on a SiN film with water or immersion oil underneath the film. d Pseudo-3D colour maps of the anomalous Raman peaks on a SiN film with water or oil underneath the film. e Pseudo-3D colour maps of PS Raman peaks on a SiN film with water or oil underneath of the film. The sphere structures were observed in all the cases. f Raman spectra of sphere peripheries normalized by PS peaks of  $1,006\text{ cm}^{-1}$  of the sphere centre on a SiN film with air, water or oil underneath the film. The anomalous periodic Raman spectra are seen with the air underneath the film (red, with air in c and d left and blue, with removal of water in c and d the third from the left). With water underneath the film, the anomalous Raman spectra decreased (purple, with water, in c and d the second from the left). With oil underneath the film, the Raman spectra indicated only the PS spectrum (green, with oil, in c and d right). g Normalized Raman spectra of sphere centre by PS peaks of  $1,006\text{ cm}^{-1}$  at the sphere centre on a SiN film with air, water or oil underneath the film. The Raman spectra under any condition show only the PS spectrum. h Relationship between the anomalous Raman peaks around the spheres and the SiN film with air, water or oil underneath the film. Six sphere samples were measured under each condition. Scale bars, 10  $\mu\text{m}$  in a, 3  $\mu\text{m}$  in b and d.

## Figure 5

Sphere size and circulating light intensity. a Averaged Raman spectra of the circumference of PS spheres of 2-7  $\mu\text{m}$  diameter on a 50 nm thick SiN film. b Relationship between the anomalous Raman peaks and sphere diameter. Six spheres were measured at each diameter and the ratio of the maximal anomalous Raman intensity to the PS peak ( $1,006\text{ cm}^{-1}$ ) at the sphere centre was calculated and normalised. The anomalous Raman peak intensity was the highest at the sphere diameter of 3  $\mu\text{m}$  but was very low at 2  $\mu\text{m}$  and 5-7  $\mu\text{m}$ . c Pseudo-3D colour maps of the PS peak and the circumferential peak for sphere diameter of 2-7  $\mu\text{m}$ . The maps of the PS peak show similar dome-like shape for all diameters (bottom row). With the anomalous Raman peak, the ring shape around the spheres was clearly observed at the SiN thickness of 3 and 4  $\mu\text{m}$  (top row). d Normalized Raman spectrum of the sphere circumference with a diameter of 2-7  $\mu\text{m}$ . The arrows in the figure indicate the peaks of each spectral periodicity. With the sphere diameter increment from 2 to 7  $\mu\text{m}$ , the periodicity of the Raman spectrum increases from 4 to 11. e Schematic of the interference produced by light circulating around the sphere. A photon travelling around the periphery of the sphere interferes with other photons while they travel around the sphere. f Raman spectrum of sphere periphery by the numerical model of equation (1). The sphere diameters of the model were set to the values where the period and phase were in close correspondence with the experimental results (left side diameters in each plot). Scale bar, 3  $\mu\text{m}$  in c.

## Figure 6

Comparison of PS spheres attached to glass and SiN film by fluorescence microscopy. a OM image of the 3  $\mu\text{m}$  diameter PS spheres on a 50 nm thick SiN film. b-d Images of spheres attached to the SiN film using three sets of fluorescence filters with excitation at 365 nm and fluorescence at 420 nm (Ex365/F420), excitation at 480 nm and fluorescence at 520 nm (Ex480/F520) and excitation at 565 nm and fluorescence at 620 nm (Ex565/F620). The PS spheres attached to the SiN film were clearly observed with all three sets of filters. e OM image of the 3  $\mu\text{m}$  diameter spheres attached to the slide glass. f-h Images of spheres attached to the slide glass using three sets of fluorescence filters with excitation at Ex365/F420, Ex480/F520 and Ex565/F520. The spheres attached to the glass showed very weak autofluorescence with a set of filters of Ex365/F420, and the sphere structure was not discerned with set of filters of Ex480/F520 or Ex565/F620. i Magnified OM image and fluorescence filter images of spheres on the SiN film indicated by a white arrow in b. In all fluorescence filter images, ring-like structures were observed around the periphery of the spheres. j Magnified OM image and fluorescence filter images of the spheres attached to the slide glass indicated by a white arrow in f. k Comparison of the line plots at the centre of the sphere image using the set of filters, Ex365/F420, on the SiN film and glass. The red, blue and green lines correspond to the RGB intensity values at the white dotted line position at the sphere centre, respectively (dotted line in i and j Ex365). The normalised RGB intensities of PS sphere on a SiN film show sharp peaks at the sphere ends (left plot). On the other hand, for the spheres on the slide glass,

the blue line shows a dome shape, but the red and green intensity show no spherical structure but just noisy (right plot). Scale bars, 10  $\mu\text{m}$  in a and 3  $\mu\text{m}$  in j.

## Figure 7

Schematic of light circulating around a dielectric microsphere. The amplitude width of the incident photon to the dielectric sphere is  $d$ , and it is assumed that half of it extends into the inside of the sphere. If the diameter of the sphere is  $D_s$ , the innermost radius of the light circling the sphere is  $1/2(D_s - d)$ . On the other hand, the outermost radius of the light circling the sphere is  $1/2(D_s + d)$ .

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