Enhanced coupling to whispering gallery modes by two-photon absorption induced by a highly focused field

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The authors report on enhanced coupling to whispering gallery modes in fluorescent polystyrene microspheres using two-photon absorption induced by a highly focused field. Due to the highly confined excitation nature under focused evanescent illumination achieved by a circularly obstructed beam, the whispering gallery modes can be excited within a small volume near the perimeter of the microsphere. As a result, the visibility, the Q factor, and the degree of polarization of the fluorescence spectra induced in the microsphere are enhanced by 60%, 37%, and five times, respectively. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2397036]

Whispering gallery mode (WGM) resonance or morphology dependent resonance (MDR) occurs when light within a dielectric microsphere resonator, which has a higher refractive index than its surroundings, gets total internally reflected and interferes.[1] Coupling the light into WGM can lead to many useful applications[2] including enhanced spontaneous emission,[2–4] structure fluorescence resonance,[5] microcavity lasing,[6] and particle-trapping microscopy.[7,8] In these applications, an effective coupling condition, that is, the effective overlapping of the excitation beam and the region where the WGMs are generated, has been achieved by a focused beam[5] or a prism.[9]

In the case of focused beam coupling to a microsphere, the three-dimensional confining nature of the focal spot is a critical factor for effective coupling. Such a confinement can be enhanced if a two-photon excitation method, in which two incident photons are simultaneously absorbed in the focal region, is used. As a result, it has been shown that the MDR effects such as the strength and polarization of the MDR peaks were highly dependent on the excitation position.[10] However, the axial dimension of the focal spot is still approximately three times larger than that in the transverse direction. This problem does not exist in a prism coupling method, in which case, an evanescent field generated by total internal reflection on a prism[9] significantly reduces the axial dimension of the excitation. However, the evanescent wave on a prism is not confined in the transverse direction. Recently, we have demonstrated the generation of a focused evanescent spot by using a high numerical aperture (NA) objective operating under the total internal reflection condition.[11–14] The aim of this letter is to demonstrate the effective coupling of WGM and the fluorescence excited by a focused evanescent field. Since the strength of the focused evanescent focus is strong, it can be used to induce two-photon excitation[11,12] which leads to a further reduction of the excitation volume. Here, we demonstrate that such an extremely tight evanescent field when used to excite WGM by two-photon-induced fluorescence is beneficial in enhancing the quality and visibility of WGM.

The geometry of focused evanescent wave coupling to WGM is similar to the design of scanning total internal reflection fluorescence microscopy.[11,12] In this geometry, the evanescent field is generated by the total internal reflection of a focused annular beam at the cover glass-air interface. The size of the annulus is chosen such that the minimum angle of convergence is equal to or greater than the critical angle for the given pair of media, thus blocking all the propagating components and generating a pure evanescent field. Such an annular beam is produced by inserting a coaxial obstruction disk, whose size is defined as ε, which is the ratio of the disk diameter D_{obs}, to the objective entrance aperture diameter D_{aperture}. By using a ring beam and focusing it using a high numerical aperture objective, a tightly confined and enhanced evanescent field is produced, with which it is possible to effectively induce two-photon absorption in a fluorescent microsphere on the cover glass, as shown in Fig. 1(a).[12] In fact, the volume of the focused evanescent spot is also dependent on the NA of the focusing objective.

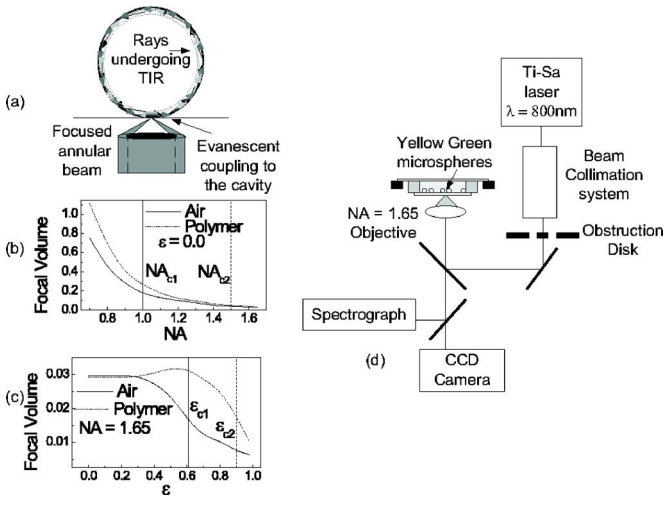

FIG. 1. (Color online) (a) Generation of a focused evanescent wave using a circular obstruction disk. Focal volume under two-photon excitation at a wavelength of 800 nm as a function of (b) NA (for $\varepsilon=0$) and (c) ε (for NA=1.65). The solid and dashed curves represent the focal volumes calculated for the glass-air and glass-polymer interfaces respectively. The refractive indices of glass, polymer, and air are 1.78, 1.5, and 1.0, respectively. (d) Schematic diagram of the experimental setup.

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Figures 1(b) and 1(c) show the focal volume ΔV under two-photon excitation as a function of NA and ε, respectively, for both the glass-air and glass-polymer interfaces, as calculated by the vectorial theory for a high NA objective.[12] It is clear that ΔV decreases appreciably as NA becomes large. In particular, ΔV decreases almost exponentially with NA when an evanescent wave is generated as NA is larger than the critical value of $NA_{c1}=1.0$ corresponding to the glass-air interface (solid curve) and for $NA_{c2}=1.5$ (dashed curve) corresponding to the glass-polymer interface. For an objective of NA=1.65, ΔV decreases drastically as soon as ε is larger than the critical value of $\varepsilon_{c1}=0.61$, determined by the critical angle at the glass-air interface and at $\varepsilon_{c2}=0.9$, determined by the critical angle at the glass-polymer interface. The slight increase in the focal volume before the critical obstruction disk size ε_{c2} is caused by the stronger effect of the propagating component of the wave after the glass-polymer interface, than the evanescent one. The reduction of ΔV between $\varepsilon=0$ and $\varepsilon=0.98$ is as large as six times in both cases because the illumination field becomes purely evanescent.

The schematic diagram of the experimental setup for coupling between two-photon fluorescence and WGM under focused evanescent wave illumination is illustrated in Fig. 1(c). A titanium-sapphire ultrashort-pulsed laser (Spectra-Physics Tsunami) at a wavelength of 800 nm and at a repetition rate of 80 MHz was used to induce two-photon absorption in polymer microspheres.[12] Fluorescent polystyrene microspheres (Polysciences Inc., Catalog No. 18140) of diameter 10 μm with an excitation peak at 486 nm were used so as to ensure efficient two-photon absorption. The pulsed laser beam was obstructed in the center using a suitable obstruction disk. A normalized obstruction disk size (ε) of 0.61 corresponded to the critical size to cut off the cone of rays below the critical angle at the cover glass-air interface. Such an annular beam was tightly focused using a high NA objective (Olympus APO 100×/NA=1.65) and made to undergo total internal reflection generating a focused evanescent field. WGMs were excited for ε of values ranging from 0 to 0.98 so that the relative coupling efficiency of the evanescent field with respect to far-field illumination could be analyzed. The strength of the fluorescence emission measured as a function of the input power, which demonstrates the quadratic nature of the multiphoton absorption, is shown in Fig. 2(a).

In order to analyze the coupling efficiency of the evanescent illumination, we first compare the two-photon fluorescence spectra recorded with NA=1.65 [Fig. 2(b)] and with the effective NA of 0.7 [Fig. 2(c)]. By reducing the aperture size and cutting off the high angle rays, the evanescent component in the illumination no longer exists, which corresponds to far-field illumination. The resonant peak at 513 nm (indicated by the arrow) for NA=0.7 becomes less pronounced compared with that with NA=1.65. The dependence of the visibility of the spectra $[V=(I_{peak}-I_{background})/(I_{peak}+I_{background})]$, where I_{peak} and $I_{background}$ are the intensities of the fluorescence peak and background, respectively] and the cavity quality factor, which is a measure of the photon storage time in the cavity[6] ($Q=\nu/\Delta\nu$, where ν is the emission frequency), on the NA is depicted in Fig. 2(d). It is seen that V and Q are increased by 71% and 40% as the effective NA is changed from 0.7 to 1.65, which confirms that the reduced focal volume, as shown in Fig. 1(b), leads to an effective coupling between the fluorescence and the WGM.

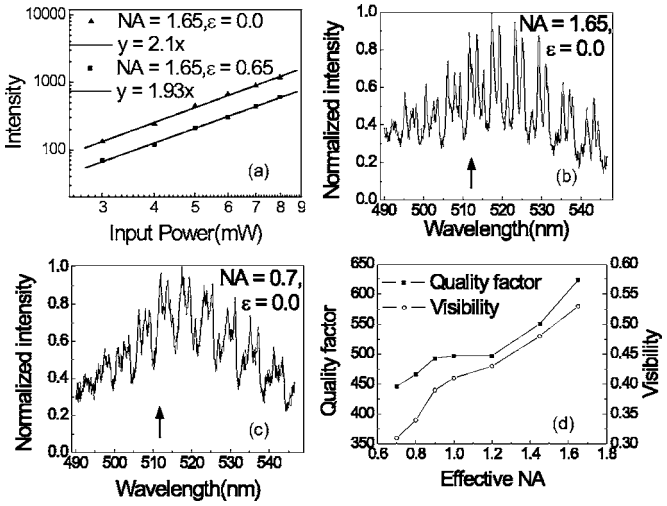

FIG. 2. (a) Logarithm plot of the fluorescence peak intensity as a function of the logarithm of the input power, where the fitted straight lines $y=2.1x$ and $y=1.96x$ give the quadratic nature of the two-photon absorption process for $\varepsilon=0$ and $\varepsilon=0.98$, respectively. [(b) and (c)] The two-photon fluorescence spectra for $\varepsilon=0$ with NA=1.65 and with NA=0.7, respectively. (d) Spectrum visibility (V) and cavity Q factor of the peak at 513 nm as a function of NA.

Such coupling can be further enhanced if the obstruction size ε is larger than the critical size, as shown in Figs. 3(a)–3(c). The two-photon fluorescence spectra from the WGM induced by the pure evanescent field illumination demonstrate that when ε increases the MDR becomes more pronounced, which is not only reflected by the increase in the ratio between the resonance peak intensity and the background but also by the decrease in the width of the MDR peaks [Figs. 3(a)–3(c)]. The visibility of the spectra (V) and the cavity quality factor (Q) measured as a function of ε are shown in Fig. 3(d), showing a significant increase when ε is larger then the critical size. For an extremely thin annulus ($\varepsilon=0.98$), an increase of 60% in the visibility and 37% in the Q factor is observed as compared with those for $\varepsilon=0$. The physical reasons for this enhancement are of two folds. First, the focal depth, which is approximately 80 nm for $\varepsilon=0.98$,[14]

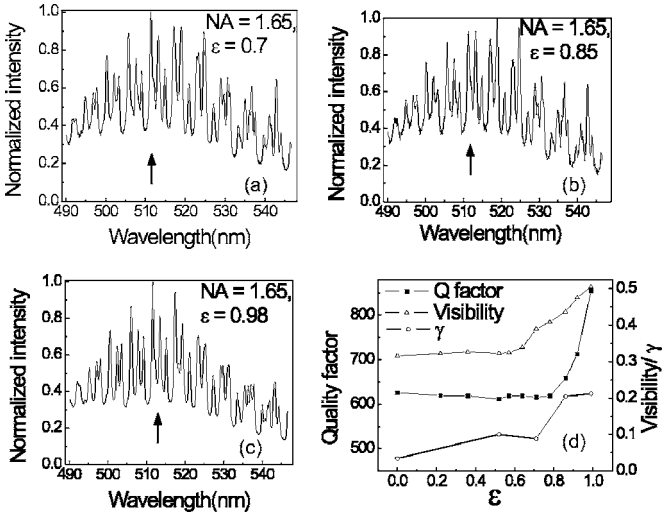

FIG. 3. [(a)–(c)] Two-photon fluorescence spectra for $\varepsilon=0.7$, $\varepsilon=0.85$, and $\varepsilon=0.98$, respectively. The arrow indicates the WGM peak at 513 nm. (d) Spectrum visibility (V), cavity Q factor, and degree of polarization (γ) of the peak at 513 nm as a function of the obstruction disk size ε.

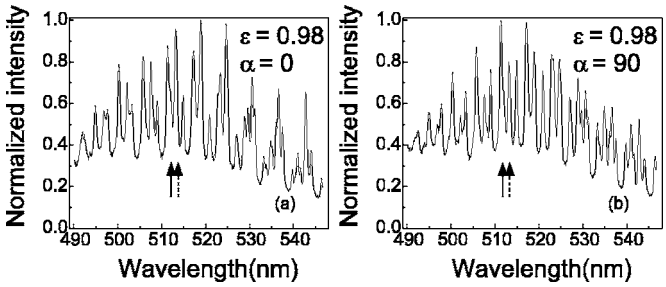

FIG. 4. Two-photon fluorescence spectra for NA=1.65 and $\varepsilon=0.98$ and corresponding to analyzer angles of (a) $\alpha=0^{\circ}$ and (b) $\alpha=90^{\circ}$. The double arrow indicates the two WGM peaks at 513 and 514 nm, respectively.

is much smaller than the cavity thickness of WGM, which is approximately 0.8 μm for a microsphere of diameter 10 μm. The excitation focal volume of the fluorescence as shown in Fig. 1(b) is well within the WGM cavity. Therefore, enhanced effective coupling is expected. Further, the component of the wave vector parallel to the interface, i.e., k_x, for a beam linearly polarized along the x direction, should contribute to stronger coupling between the fluorescence and WGM within the microsphere.

The degree of polarization of the individual peaks was also investigated by placing an analyzer in the detection arm. Figures 4(a) and 4(b) show the evanescent wave excited spectra with an analyzer at angles of $\alpha=0°$ and 90° with respect to the incident polarization direction. It is noted that the relative strengths of the two peaks indicated by the two arrows change with the analyzer position. When the analyzer was rotated by 90° the peak at 514 nm (indicated by dashed arrow) became less pronounced while the peak at 513 nm (indicated by solid arrow) became stronger. This demonstrates that the MDR peaks are polarized and that the peaks at 513 and 514 nm have orthogonal polarization states representing the TE and TM modes in a microcavity, which is consistent with the previous report elsewhere.[7] To quantify this phenomenon, we measured the degree of polarization γ [defined as $\gamma=(I_{\alpha=\max}-I_{\alpha=90°})/(I_{\alpha=\max}+I_{\alpha=90°})$, where $I_{\alpha=\max}$ and $I_{\alpha=90°}$ represent the maximum intensity when the analyzer is placed at 0° and 90°, respectively] for $\varepsilon=0$ and $\varepsilon=0.98$, showing an increase in γ by five times. This feature further indicates that the highly localized evanescent field provides strong coupling of the fluorescence to WGM.

To summarize, we have demonstrated enhanced coupling of two-photon-induced fluorescence to WGM by using a highly focused wave, because of the effective overlapping between the fluorescence volume and the WGM cavity. Such an enhancement leads to a significant increase in the visibility, the Q factor, and the degree of polarization if a purely evanescent focal spot produced by using an annular high NA objective is adopted. The use of such an efficiently excited optical microcavity would provide resonance sensing mechanisms in near-field optics.

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