Observation of orthogonally polarized transverse electric and transverse magnetic oscillation modes in a microcavity excited by localized two-photon absorption

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We report on the observation of orthogonally polarized transverse electric (TE) and transverse magnetic (TM) oscillation modes in a microcavity excited by localized two-photon absorption. The polarization-dependent features of morphology-dependent resonance (MDR) effects in a microsphere under two-photon fluorescence excitation are quantitatively investigated. In addition to a clear separation of excitation and resonance wavelengths under two-photon excitation, the fluorescence emission can be tightly controlled in three-dimensional space within a microsphere. The experimental results demonstrate not only the orthogonal polarization nature of TE and TM oscillation modes but also the dependence of the strength and the polarization properties of MDR peaks on excitation locations in a microsphere. © 2002 American Institute of Physics. [DOI: 10.1063/1.1531222]

Morphology-dependent resonance (MDR) arises due to the constructive interference of light rays at near-glancing angles by total internal reflection within a microcavity. A dielectric sphere possesses natural internal modes of oscillation at characteristic frequencies corresponding to the specific ratio of size to wavelength. These modes of oscillation are known as whispering gallery modes.1 The MDR effect was first discussed by Purcell,2 who noted that the changes in the final density of states per unit volume and unit frequency would lead to a greatly enhanced probability of spontaneous emission over that normally observed in free space. MDR effect has also been demonstrated by the enhancement of the fluorescence spectrum at particular wavelengths with a microsphere.3–5

Although the MDR effect and its subsequent applications, such as microcavity lasing, have been investigated for microparticles, microdroplets and other microobjects,6–9 it has been difficult in the past to quantitatively determine the features, such as the state of polarization, of MDR peaks representing transverse electric (TE) and transverse magnetic (TM) modes. Due to the difficulty of confining the excitation volume in three-dimensional (3D) space under single-photon excitation, previous experiments have been based on an averaged effect over a large excitation volume, despite the fact that the characteristics of individual MDR peaks depend on the location of the excitation spot in a microobject. With the rapid development of ultrashort pulse lasers in the past two decades, multi-photon fluorescence excitation becomes available in many research laboratories.10–12 The advantage of multi-photon fluorescence excitation over single-photon excitation is that a highly confined excitation volume in 3D space can be achieved due to the nonlinear intensity response of multi-photon excitation.10 In addition, a large separation of excitation and resonance wavelengths is inherent with this technique.

In this letter we demonstrate the polarization-dependent features of MDR peaks in a microsphere under two-photon excitation. Due to the highly localized spatial nature of two-photon excitation, the introduction of MDR in a microcavity can be tightly controlled. Therefore fluorescence excitation at various spots in 3D space of a microsphere can be investigated in detail.

The experiment system layout is shown in Fig. 1(a). A train of linearly polarized 80 fs pulses of 870 nm wavelength (Spectra-Physics MaiTai) is coupled directly into a scanning microscope (Olympus FV300 IX). The polarization direction of the laser is along the x direction shown in Fig. 2. A high numerical aperture (NA=1.2) water immersion objective

![Image of experimental setup](https://example.com/image1)

FIG. 1. (a) Schematic diagram of the experimental setup; (b) fluorescence spectrum (r=a, θ=0); (c) fluorescence intensity as a function of input power (r=a, θ=0).
The fluorescence emission from an excited microsphere is illustrated in Fig. 1. The fluorescence light is truly excited under two-photon absorption and/or scattering. A polarization analyzer is put in the detection path in order to investigate the polarization nature of the MDR peaks periodically change with the analyzer rotation angle. When a polarization analyzer is introduced in the detection path, the relative strength of these two MDR peaks changes [Fig. 3(b)]. If the analyzer angle \( \alpha \) rotates by \( 90^\circ \), the peak at 509.6 nm becomes less pronounced compared to the peak at 511.9 nm [Fig. 3(c)]. Rotating the analyzer by another \( 90^\circ \) leads to the similar fluorescence spectrum shown in Fig. 3(b). In order to quantify the polarization nature of the MDR peaks, the strength of the two adjacent MDR peaks as a function of the analyzer rotation angle is investigated [Fig. 3(d)]. The result shows that MDR peaks periodically change with the analyzer angle \( \alpha \); for the two adjacent peaks, one peak is maximized while the other is minimized. This result demonstrates the polarization nature of MDR peaks; the two adjacent peaks representing other is minimized. This result demonstrates the polarization nature of MDR peaks; the two adjacent peaks representing also the relative strength between different MDR peaks. For example, if the excitation spot moves along a radial direction in the equatorial plane [Figs. 2(a)–2(c), \( 0 \leq r < a, \theta = 0^\circ, \phi = 0^\circ \)], the MDR peaks are greatly enhanced when the excitation spot moves towards the equator [Fig. 2(c), \( r = a, \theta = 90^\circ, \phi = 0^\circ \)]. It is also shown that the relative strength between the adjacent MDR peaks changes if the excitation spot moves in the equatorial plane [Figs. 2(i–iii), \( 0 \leq \phi < 180^\circ, r = a, \theta = 0^\circ \)].

Among all the factors that cause the changes in fluorescence emission due to different spatial locations of excitation, the polarization nature of the MDR peaks (demonstrated in Fig. 3) plays an important role. For a focal spot on the equator of a microsphere (\( r = a, \theta = 0^\circ \)), the emission spectrum detected without an analyzer is shown Fig. 3(a). Let us pay particular attention to two adjacent MDR peaks at wavelengths 509.6 and 511.9 nm, marked by solid and dashed arrows, respectively. When a polarization analyzer is introduced in the detection path, the relative strength of these two MDR peaks changes [Fig. 3(b)]. If the analyzer angle \( \alpha \) rotates by \( 90^\circ \), the peak at 509.6 nm becomes less pronounced compared to the peak at 511.9 nm [Fig. 3(c)]. Rotating the analyzer by another \( 90^\circ \) leads to the similar fluorescence spectrum shown in Fig. 3(b). In order to quantify the polarization nature of the MDR peaks, the strength of the two adjacent MDR peaks as a function of the analyzer rotation angle is investigated [Fig. 3(d)]. The result shows that MDR peaks periodically change with the analyzer angle \( \alpha \); for the two adjacent peaks, one peak is maximized while the other is minimized. This result demonstrates the polarization nature of MDR peaks; the two adjacent peaks representing TE and TM oscillation modes of a microcavity have orthogonal polarization states. It is also noticed that the measured separation of the two adjacent MDR peaks of same polarization is approximately 5.9–6.0 nm around the wavelength 510 nm. For a polymer microsphere of refractive index \( n = 1.59 \), it agrees well with the result of 6.0 nm estimated by

\[
\Delta \lambda = \frac{2}{\pi \alpha^2} \frac{\lambda^2}{\sqrt{n^2 - 1}} \arctan \left( \sqrt{\frac{n^2 - 1}{n^2 - 1}} \right) \tag{1}
\]

based on the plane-wave Mie scattering theory.\(^{14}\)
Due to the highly localized nature of two-photon absorption, the characterization and manipulation of the strength of individual MDR peaks within 3D space of a cavity can be achieved. In order to quantify the MDR strength and its polarization nature in relation to the excitation position, we introduce two measurable quantities, visibility \([V = (I_{\text{peak}} - I_{\text{background}})/(I_{\text{peak}} + I_{\text{background}})]\) and the degree of polarization \([\gamma = (I_{a=\text{max}} - I_{a=90})/(I_{a=\text{max}} + I_{a=90'})]\), where \(I_{\text{peak}}\) and \(I_{\text{background}}\) are the intensity of MDR peaks and the background fluorescence, respectively. \(I_{a=\text{max}}\) and \(I_{a=90'}\) represent the maximum intensity of a peak and the intensity of the peak when the analyzer rotated by \(90^\circ\).

The visibility and the degree of polarization as a function of polar coordinates \((r, \theta, \phi)\) are demonstrated in Fig. 4. Due to the photobleaching effect associated with high power illumination, the results along each spherical coordinate are obtained for a given microsphere. In the radial direction \((0 \leq r \leq a, \theta = 0^\circ, \phi = 0^\circ)\), an increase from 3.5% to 48% in visibility between the center of the cavity and the perimeter is evident [Fig. 4(a)]. It is intuitive that the localization of a focal spot at the perimeter leads to an increase in the coupling of the resonance rays due to the glancing angles of incidence with respect to the boundary. A similar trend is also observed in the degree of polarization [Fig. 4(d)], due to the fact that the MDR peaks are highly polarized and the background fluorescence is unpolarized.

The dependence of the MDR visibility on the excitation positions in the meridian plane \((-90^\circ \leq \theta \leq 90^\circ, r = a, \phi = 0^\circ)\) is shown in Fig. 4(b). The MDR effect becomes more pronounced when the incident illumination is localized around the equatorial plane of the microcavity, i.e., when \(\theta = 0^\circ\). It is because more rays which are coupled into the cavity satisfy the total internal reflection condition if the focal spot is at the equator of a sphere. The break in symmetry between the two hemispheres can be ascribed to the multiple reflection between the cavity and the substrate cover slip when \(\theta = -90^\circ\) and to the spherical aberration induced by focusing through the sphere when \(\theta = 90^\circ\). With the highly polarized MDR peaks and the unpolarized background fluorescence, a similar trend is observed in terms of degree of polarization [Fig. 4(e)].

The visibility and the degree of polarization as a function of azimuth angles for excitation locations in the equatorial plane \((0 \leq \phi \leq 360^\circ, \theta = 0^\circ, r = a)\) are shown in Figs. 4(c) and 4(f), respectively. It is found that a periodic variation in the peak visibility is due to the excitation position. When the polarization of the incident light is fixed, the strength of the MDR peak can be controlled by the focal position. This occurs due to the fact that the polarization state of the incident beam with respect to the boundary of the microcavity changes with different \(\phi\) values. For example, if the excitation beam is linearly polarized in the \(x\) direction (as indicated in Fig. 2), the polarization direction is parallel to the surface of the sphere for excitation locations i and iii, and becomes perpendicular for excitation location ii. However, it is also noted that the degree of polarization in the equatorial plane remains constant. This result suggests that the maximum strength of MDR peaks does not change with different focal positions on the equator, however the polarization direction of MDR peaks shifts as the result of changing polarization direction of the excitation.

In conclusion, we have demonstrated that MDR effects are highly dependent on the excitation location in a microsphere. With two-photon excitation, the fluorescence emission can be tightly controlled in 3D space within a microsphere, which allows quantitative characterization of the strength and polarization nature of MDR peaks. It has been demonstrated that two adjacent peaks representing TE and TM oscillation modes of a microcavity have orthogonal polarization states. With its intrinsic advantages of 3D confined excitation volume and the clear separation of the excitation and emission wavelengths, two-photon microscopy provides us with a unique tool to gain fundamental knowledge about a microcavity.

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2. E. M. Purcell, Phys. Rev. 69, 681 (1946).