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著者	宮嵜　博司
journal or publication title	Physical review letters
volume	82
number	23
page range	4623-4626
year	1999
URL	http://hdl.handle.net/10097/35135

doi: 10.1103/PhysRevLett.82.4623

Tight-Binding Photonic Molecule Modes of Resonant Bispheres

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(Received 9 November 1998)

We observe coherent resonant coupling of optical whispering-gallery modes in fluorescence from dye doped polymer bispheres with diameters ranging from 2 to 5 μm. By monitoring the frequencies of fluorescence peaks of individual spheres, we sort out two spheres with appropriate size matching and bring them into contact. Wave optics calculation also gives good agreement with the experiment. By taking into account harmonic coupling of the whispering-gallery modes, the obtained features of normal mode splitting are well explained by the tight-binding photon picture. [S0031-9007(99)09349-7]

PACS numbers: 42.60.Da

Manipulation of light path in micrometer length scale has recently attracted considerable attention from both fundamental and application points of view. Conventionally, the manipulation is based on the photonic crystal concept [1–3]. In photonic crystals, which have periodic modulation of the refractive index, propagation of the light wave is governed by a weak potential. Correspondingly, such an approach can be referred to as a nearly free photon approach analogous to the nearly free electron approach in band theory. Alternatively the micromanipulation of light can be achieved by exploring the possibility of confining the light in a small unit of the wavelength size. Light propagates through the system of such units due to the coupling between the nearest neighbors. This approach is referred to as the tight-binding photon approach [4]. Within the tight-binding photon approach we can guide the optical waves by connecting the units in the arbitrarily shaped microstructures.

The microspheres are the most natural choice of the unit to be employed in the tight-binding photon device. It is known that a dielectric sphere acts as a unique optical microcavity which has very long photon storage time within a small mode volume [5–8]. In particular, Q factors of the order of 10^{10} have been observed for whispering gallery modes (WGM's) in quartz spheres with a diameter of several tens of micrometers [9–13], and the mode structure of a pair of these large spheres in contact has been studied [14]. However, in order to explore the feasibility of micromanipulation of light, one has to confirm the existence of the coherent coupling between spheres of the size of a few times of optical wavelength. Lorenz-Mie theory predicts long photon lifetime even for small spheres, giving, for example, nearly 30 ps for a 4 μm sphere with a refractive index of 1.59. This has allowed one to propose such relatively small spheres to be employed as "photonic atoms" [15] for the tight-binding scheme. However, the coherent coupling between two adjacent microspheres of such size range have not been realized until now. The coherent coupling results in the splitting of the corresponding WGM's and is a manifestation of the well-known phenomena of the normal mode splitting (NMS) in coupled harmonic oscillators. However, although some attempts have been made [16], NMS has not yet been observed because of the difficulty in the precise size control of the spheres.

In this Letter, we report on the observation of normal mode splitting in the system of two polymer spheres in contact (bisphere) under extreme size control. We study bispheres formed from the spheres of diameters ranging from 2 to 5 μm. The sufficiently narrow linewidth and wide separation of WGM's in this size range allow us to avoid intricate band mixing. The frequencies of the observed bisphere resonances agree with the wave optics calculations. In order to examine the feasibility of tight-binding manipulation of light waves in a structure composed of connected microspheres, we estimate an intersphere coupling constant which is found to be larger than the WGM linewidth.

We use monodispersive polystyrene spheres (refractive index 1.59) which are soaked with a solution of dye (Nile Red, concentration is about 10^{-2} mol/l, fluorescence FWHM is about 70 nm). Dye doped spheres are sown on a glass plate under a microscope and can be manipulated with an optical fiber probe which is made by pulling the multimode optical fiber in a frame. By using a microscope objective lens, the individual sphere is excited with the second harmonics of a cw Q-switch Nd-YLF laser ($\lambda = 527$ nm). The dye fluorescence is collected with the same fiber probe and sent to a spectrometer with a liquid nitrogen cooled CCD (charge-coupled device) detector. We use both 50 cm and 1 m focal length spectrometers with spectral resolution of 0.06 and 0.03 nm, respectively.

Figure 1 shows an example of fluorescence spectrum from the individual sphere of about 4.1μm in diameter. Sharp lines, which originate from the strong modification

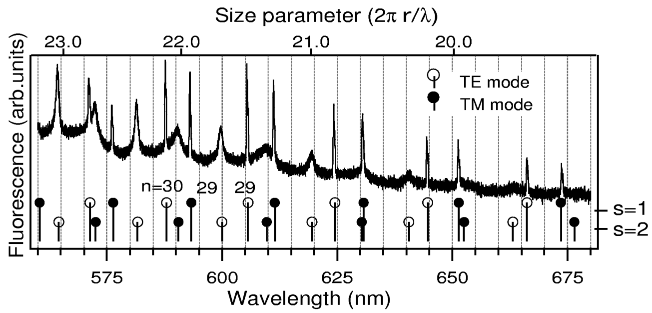

FIG. 1. Typical fluorescence spectrum from 4.1 μm sphere. Calculated positions for $s = 1$ and $s = 2$ are shown at the bottom of the figure.

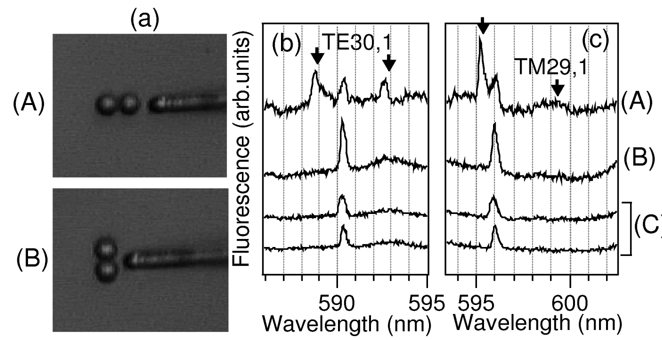

FIG. 2. (a) Microscope images of the bisphere and the fiber probe. Diameter of the probe is about 5 to 10 μm. We detect the emission in two different geometries; one is parallel configuration (A), the other is perpendicular configuration (B). (b),(c) Spectra of resonance bisphere of TE30,1 mode and TM29,1 mode in parallel configuration (A) and perpendicular configuration (B). Spectra (C) show the fluorescence of individual spheres before contact. Two spheres have almost the same diameter. The arrows indicate the coupled modes.

of the vacuum field inside the sphere [6–8,17–19], appear at the resonance frequencies of WGM's with TE and TM polarizations.

According to the Lorenz-Mie theory, each WGM can be specified by the mode number n (indicating the number of light wavelengths around the circumference), the order number s (corresponding to the number of maxima in the radial dependence of the internal electric field), and the azimuthal mode number m (giving the orientation of the WGM's orbital plane). Since the frequency of WGM does not depend on m, each resonance has $(2n + 1)$-fold degeneracy.

For spheres of diameter from 2 to 5 μm we can identify the mode indices n and s of the high-Q WGM by comparing the positions of the lines in the fluorescence spectra with the predictions of the Lorenz-Mie theory. The widths of the resonances in the fluorescence spectrum give the actual Q factor of the particular sphere. Once we label the fluorescence lines with the mode indices n and s, we can judge the relative size of the sphere within an error of $1/Q$ which is about 0.05% for a 5 μm sphere. Spheres of 2 μm in diameter are found to have a Q factor of the order of 10^2. This is comparable with the radiative leakage rate which follows from the Mie theory. Spheres of diameters more than 4 μm are found to have Q factors of the order of 10^3. This is 2 orders less than the radiative leakage rate [20].

In order to make the bisphere, we choose two spheres of the desired size by comparing the frequencies of the specific WGM resonances. By handling the optical fiber probe attached to the three axis mechanical stage, we pick up one sphere and attach it to the other.

We measure the bisphere fluorescence in parallel configuration [Fig. 2(a)-(A); the fiber probe is set parallel to the axis of the bisphere] and perpendicular configuration [Fig. 2(a)-(B); the fiber probe is set perpendicular to the axis of the bisphere]. Figures 2(b) and 2(c) show the fluorescence spectra of individual spheres (C) and the bisphere (A, B) in the vicinity of TE30,1 (TE mode of $n = 30$ and $s = 1$) and TM29,1 (TM mode of $n = 29$ and $s = 1$) resonances. For both the polarizations, we observe new peaks due to the intersphere coupling, in the parallel configuration (A) but not in the perpendicular configuration (B). To explain this, one should recall that each WGM of a given n and s has $(2n + 1)$-fold degeneracy associated with the azimuthal mode number m. Hence $2n + 1$ orientations of the WGM orbital plane are permitted and, correspondingly, $(2n + 1)^2$ combinations of the intersphere couplings are generally possible. However, the intersphere coupling is maximum for the pair of modes whose orbitals include the contact point and lay in the same plane. Therefore, the fluorescence of the coupled modes is maximum in the direction parallel to the bisphere axis. As a result, the signal from the coupled intersphere modes is more pronounced in the parallel configuration than in the perpendicular configuration. As we can see in Figs. 2(b) and 2(c), the uncoupled modes fluorescence also contributes to the signal. By reducing the aperture of the fiber or keeping the probe away from the bisphere, this signal diminishes.

The dependence of the mode coupling on the orbital plane orientation breaks the degeneracy with respect to the azimuthal indices. In particular, bisphere modes originating from the coupling of the WGM's with the different combinations of azimuthal numbers contribute differently to the signal. Correspondingly, the line shape of the signal represents the energy distribution among the coupled modes. The asymmetric line shape of the coupled modes on Figs. 2(b) and 2(c) indicate that the stronger the coupling between WGM's, the more the energy in the resulting bisphere mode.

In order to study the dependence of the WGM coupling on the sphere size (size detuning), we fix the diameter of one sphere and change the diameter of the second sphere in the scale of 0.3%. Figure 3 shows the detuning dependence of the bisphere fluorescence in the vicinity of

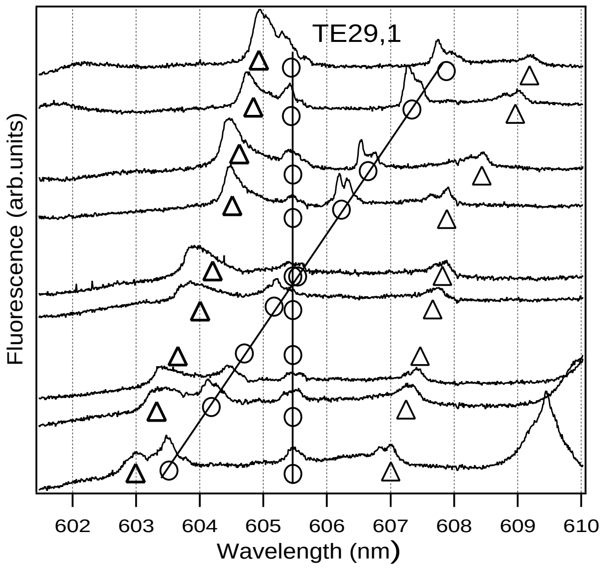

FIG. 3. Emission spectra of nine bispheres with slightly different sized spheres. Fiber probe is set parallel to the axis of the bisphere. Circles indicate the uncoupled modes. Triangles show the coupled modes of bisphere.

TE29,1 mode for various pairs with size detuning. Circles show the uncoupled original resonance, and triangles show coupled modes. We clearly see the anticrossing behavior which is the signature of NMS.

Figure 4 shows the dependence of bisphere resonance on detuning for TE30,1 and TM29,1 modes. Positions of modes before contact are shown with dashed lines. Both TE30,1 and TM29,1 modes show anticrossing with some symmetry. Such an asymmetry could be attributed to the influence of the broad second order $(s = 2)$ WGM's, and their positions are shown in the figure with thick shaded lines.

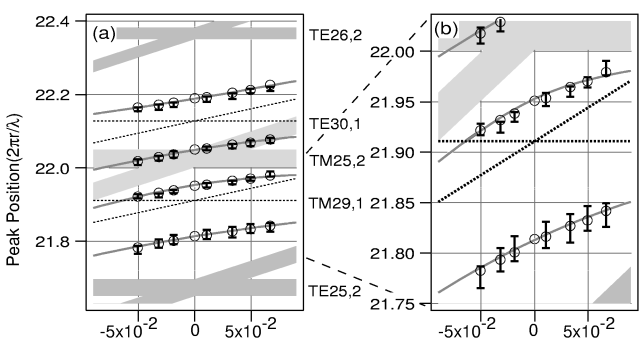

FIG. 4. Resonance frequencies of coupled TE30,1 and TM29,1 for various bispheres with different size detuning, $\Delta x \equiv 2\pi \Delta a/\lambda$. Dashed lines indicate the resonances of uncoupled modes. Large open circles show the result of wave optics calculation. Solid lines show the calculated normal modes of bisphere as a function of size detuning using the coupled harmonic oscillator model. Coupling constants are shown in Tables I and II.

The observed features of intersphere coupling can be explained by the resonance enhancement of the internal field in bispheres. By expanding the internal and external fields of each sphere in terms of the vector spherical harmonics under the plane wave excitation whose wave vector is parallel to the bisphere axis, we solve numerically the coupled linear equation by the matrix inversion method [21]. The energy of the internal field is obtained from the expansion coefficients as a function of the incident plane wave parameters and sphere radii. Excellent numerical convergence is achieved with the summation of n up to 49. The spectrum shows prominent peaks asymmetrically located on both sides of TM29,1 and TE30,1 modes. In between these prominent peaks there appears a series of less intense peaks converging to the original frequencies of WGM's. These substructures make the line shape asymmetric as we show in Figs. 2(b) and 2(c). Open circles of Fig. 4 show the positions of the prominent peaks. One may observe an excellent agreement between calculation and experiments from Fig. 4. Note that both theory and experiment show asymmetry in the mode splitting.

In order to explore the feasibility of the tight-binding scheme for the micromanipulation of light, it would be desirable to introduce a mode overlap parameter which could describe the intersphere coupling. It is clear that these parameters will be determined by the convolution of the modes' electric fields. Therefore, one can expect the strong coupling for the modes with the matching frequencies whose orbitals lay nearly in the same plane and are close to the contact point. We will refer coupling between the modes with the same and different mode numbers as "intramode" and "intermode" coupling, respectively. By introducing the phenomenological coupling parameters of the dimension of frequency, which accounts for the intermode and intramode coupling for the relevant modes, we can describe the bisphere fluorescence in standard terms of the interacting harmonic systems. In order to reproduce the experimental results we take into account the intramode coupling between the WGM's with $s = 1$, intermode coupling of the WGM's with $s = 1$ and $s = 2$ and neglect the coupling between TE and TM modes. The best fit values of the coupling parameters are shown in Table I (TE mode) and Table II (TM mode); and the corresponding detuning dependences are shown in Fig. 4 as solid lines.

One can see from Fig. 4 that this model well explains the overall features of the normal modes of bispheres and the asymmetric behavior of the upper mode branch of the TM29,1 due to the strong mixing with the TM25,2 mode. The best fit values of the coupling parameters are almost constant for all the modes involved in the narrow region of mode number variation from $n = 25$ to 30.

We perform four series of measurements using spheres with diameters of 2, 3, 4.1, and 5 μm and estimate the

TABLE I. Best fit values of coupling parameters (TE modes) normalized by resonance frequency.

	TE26, $s = 2$	TE30, $s = 1$	TE25, $s = 2$
TE26, $s = 2$	3.4×10^{-3}	3.4×10^{-3}	$\cdots$
TE30, $s = 1$	3.4×10^{-3}	3.3×10^{-3}	3.4×10^{-3}
TE25, $s = 2$	$\cdots$	3.4×10^{-3}	3.4×10^{-3}

coupling parameters. The obtained coupling parameters normalized to the resonance frequencies are 7×10^{-3} for 2 μm ($n = 14$); 5×10^{-3} for 3 μm ($n = 21$); 4×10^{-3} for 4.1 μm ($n = 29$); and 3×10^{-3} for 5 μm ($n = 36$). This implies that the coupling parameters decrease with increasing sphere diameter or mode number. To explain such a behavior, one can recall that the smaller the spheres, the more the electric field outside the spheres, and hence the coupling parameter increases. One could be reminded here that the mode number represents the order of the Hankel function which gives the field of WGM outside the sphere. In the experiment two spheres are in contact and hence we obtain the maximum available coupling for a given size. The coupling can be reduced by increasing the distance between spheres as it has been demonstrated for bigger spheres [14]. We would like to emphasize that the obtained coupling parameters are much larger than the line widths, indicating the dominance of coherent resonant coupling.

In summary, we observe the coherent coupling of WGM's in bispheres. By the precise control of the sphere size, anticrossing behavior of the mode coupling is clearly demonstrated. The features of normal mode splitting are perfectly reproduced by the wave optics calculations. Overall features of obtained bisphere normal modes are well explained with the coupled harmonic oscillator model. The observed asymmetry in the splitting of upper and lower branches is ascribed to intermode mixing of $s = 1$ mode with adjacent $s = 2$ modes. In the examined size region of spheres, the coupling parameters are found to be much larger than the line width and smaller than the mean mode separation of the WGM's. This ensures the feasibility of the tight binding manipulation of light waves in a structure composed of contacting spheres. This scheme will also bring new insight into the application of WGM's for micro-optical devices [22,23].

TABLE II. Best fit values of coupling parameters (TM modes) normalized by resonance frequency.

	TM25, $s = 2$	TM29, $s = 1$	TM24, $s = 2$
TM25, $s = 2$	2.8×10^{-3}	3.5×10^{-3}	$\cdots$
TM29, $s = 1$	3.5×10^{-3}	3.3×10^{-3}	3.5×10^{-3}
TM24, $s = 2$	$\cdots$	3.5×10^{-3}	2.8×10^{-3}

The author would like to acknowledge Dr. Yu. P. Svirko and Dr. C. Ramkumar for many fruitful discussions and Dr. Y. Li for her help during measurements. This work was supported by a grant-in-aid for Scientific Research in the Priority Area "Near-Field Nano Optics."

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[1] A. Mekis *et al.*, Phys. Rev. Lett. **77**, 3787–3789 (1996).
[2] J. D. Joannopoulos, P. R. Villeneuve, and S. Fan, Nature (London) **386**, 143–149 (1997).
[3] J. D. Joannopoulos, P. R. Villeneuve, and S. Fan, Solid State Commun. **102**, 165–173 (1997).
[4] E. Lidorikis, M. M. Sigalas, E. N. Economou, and C. M. Soukoulis, Phys. Rev. Lett. **81**, 1405–1408 (1998).
[5] P. R. Conwell, P. W. Barber, and C. K. Rushforth, J. Opt. Soc. Am. A **1**, 62–67 (1984).
[6] R. E. Benner, P. W. Barber, J. F. Owen, and R. K. Chang, Phys. Rev. Lett. **44**, 475–478 (1980).
[7] M. Kuwata-Gonokami, K. Takeda, H. Yasuda, and K. Ema, Jpn. J. Appl. Phys. **31**, L99–L101 (1992).
[8] M. Kuwata-Gonokami and K. Takeda, Opt. Mater. **9**, 12–17 (1998).
[9] V. B. Braginsky, M. L. Gorodetsky, and V. S. Ilchenko, Phys. Lett. A **137**, 393–397 (1989).
[10] L. Collot *et al.*, Europhys. Lett. **23**, 327–334 (1993).
[11] V. Sandoghdar *et al.*, Phys. Rev. A **54**, R1777–R1780 (1996).
[12] M. L. Gorodetsky, A. A. Savchenkov, and V. S. Ilchenko, Opt. Lett. **21**, 453–455 (1996).
[13] D. W. Vernooy *et al.*, Opt. Lett. **23**, 247–249 (1998).
[14] V. S. Ilchenko, M. L. Gorodetsky, and S. P. Vyatchanin, Opt. Commun. **107**, 41–48 (1994).
[15] S. Arnold *et al.*, J. Opt. Soc. Am. B **9**, 819–824 (1992).
[16] S. Arnold, A. Ghaemi, P. Hendrie, and K. A. Fuller, Opt. Lett. **19**, 156–158 (1994).
[17] A. J. Campillo, J. D. Eversole, and H.-B. Lin, Phys. Rev. Lett. **67**, 437–440 (1991).
[18] H.-B. Lin and A. J. Campillo, Phys. Rev. Lett. **73**, 2440–2443 (1994).
[19] S. Arnold, J. Chem. Phys. **106**, 8280–8282 (1997).
[20] The light absorption by dye molecules and light scattering by an imperfect surface reduce the Q factor to the order of 10^3. Nonideal spherical shape of spheres lifts the degeneracy with respect to the azimuthal mode indices, giving rise to the broadening and asymmetry of the fluorescence lines. The upper limit of the eccentricity of the spheres used in this experiment can be estimated from the width of narrowest line. For example, the narrowest line width of the 5 μm spheres is 0.4 nm which ensure the eccentricity less than 0.08%.
[21] M. Inoue and K. Ohtaka, J. Phys. Soc. Jpn. **52**, 3853–3864 (1983); K. A. Fuller, Appl. Opt. **30**, 4716–4731 (1991).
[22] X. Fan, A. Doran, and H. Wang, Appl. Phys. Lett. **73**, 3190–3192 (1998).
[23] C. Gmachl *et al.*, Science **280**, 1556–1564 (1998).