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International Frequency Sensor Association (IFSA).

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Registration	November 5, 2010
Camera ready	November 5, 2010

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Sensitivity Enhancement of Biochemical Sensors Based on Er^{+3} Doped Microsphere Coupled to an External Mirror

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Abstract: In this paper we proposed an active optical sensor designed based on the Er^{+3}-doped microsphere coupled to an external mirror. The microsphere-mirror coupling causes the degeneracy splitting of the resonance frequencies. Each of resonance frequencies splits in to two different resonance frequencies .The coupling coefficient changes as a result of altering the refractive index of surrounding medium. So, the presence of bio/chemical analytes can be detected by measuring the change of laser output power, laser frequency or the difference between frequencies of the red and blue shifted modes. In the presence of mirror at least one order of magnitude sensitivity enhancement is obtained relative to the active microsphere biochemical sensors. *Copyright © 2010 IFSA.*

Keywords: Biosensor, Erbium, External feedback, Microsphere.

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1. Introduction

The possibility of using dielectric micron size spherical particles (microspheres) as resonators has been studied since the early days of lasers [1].

A microsphere doped with a sufficient concentration of rare earth ions provides an active cavity which acts as both the active medium and the resonator for laser emission [2, 3]. Strongly temporal and spatial confinement of light in the microspheres and also their morphology-dependent resonance frequencies makes them suitable for a large number of applications such as quantum optics, nonlinear optics, photonics and bio/chemical sensing systems [4-6].

When a molecule is absorbed on the surface of a microsphere, the evanescent field of the microsphere mode polarizes the attached molecule. When the refractive index of the surrounding medium of the microsphere resonator (or microsphere laser) changes, the resonance frequencies of the microsphere (or oscillation frequencies of the microsphere laser) changes. These changes in resonance frequency or oscillation frequency due to the change of the refractive index of the surrounding medium lay the foundation of micro cavity optical sensors.

High quality factor micro cavities such as microspheres [3, 7], microtoroids [2, 8] and microrings [9] on the basis of Whispering Gallery Mode frequency shift have already been proposed and demonstrated for biosensor applications. To improve the sensitivity of the optical biosensor a design on the basis of active microsphere is also proposed previously [10]. In this paper, sensitivity enhancement of a biosensor based on the active microsphere with external feedback is proposed. The external cavity is made of a metallic or non metallic mirror. It is shown that the output power and frequency of oscillation are strongly dependent on the optical path length between the microsphere and external mirror surface. The change of refractive index of surrounding medium causes to change the coupling coefficient between the microsphere and external mirror, hence, the laser output power and laser frequencies are changed. Depending on the region of laser operation, several methods for detecting biochemical analytes exist. For the single mode region of operation, the change of laser output power or laser frequency signifies the presence of biochemical analytes. In the bi-mode region of operation of the microsphere laser, the frequency difference between the even and odd modes is a suitable measure for the microsphere biochemical sensor.

This paper is organized as follows: Oscillation frequencies are determined in section 2. Calculation of the sensitivity enhancement of proposed biochemical sensor is presented in section 3. Finally, the paper is enclosed with some conclusion in section 4.

2. Resonance and Oscillation Frequencies

A microsphere doped with optically active erbium ions shows lasing at the ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+} around 1.5 µm [3, 11]. If one places this microsphere near an infinite flat mirror, the resonance frequencies of the microsphere and hence the lasing frequencies will change.

To find the sensitivity of the proposed biosensor, it is necessary to obtain variation of oscillation frequency of the microsphere laser near an infinite mirror plate versus their coupling coefficient. An input pump power is coupled to the microsphere through a full-tapered fiber and the laser power couples to the output through the same fiber (Fig. 1). In this analysis, it is assumed that both the pump and laser modes are coincident with one of the fundamental modes of the mirror-micro resonator system. By this assumption the mode-pulling effect is negligible.

To maintain the electric field perpendicular to the mirror surface, the non mirror may be replaced by the image of the microsphere in the mirror. The image is shown in the right hand side of Fig. 2.

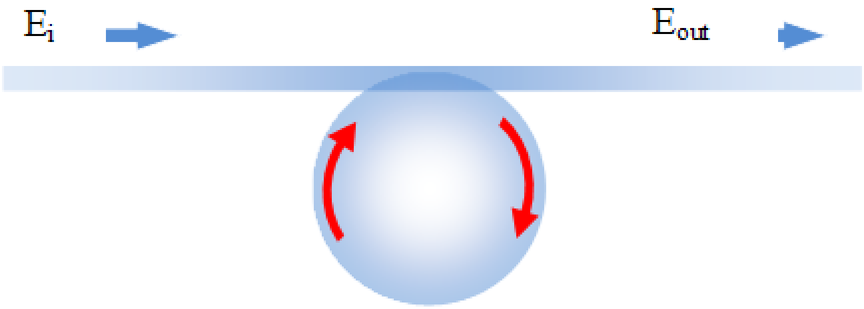


Fig. 1. The full tapered fiber couples to the microsphere.

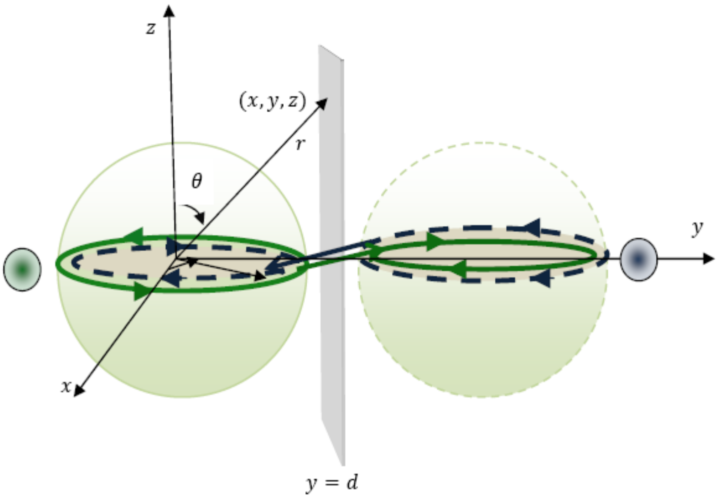


Fig. 2. The microsphere and external feedback mirror, that the mirror may be replaced by the image of the microsphere in the mirror.

Almost of the theories developed for computing the resonance frequencies of dielectric microspheres on or near a plane of infinite conductivity are based on the Mie scattering [12, 13]. In this paper, the transfer matrix method [14, 15] is employed for prediction of the lasing frequency and laser output power versus the coupling coefficient or optical path length between the microsphere and the mirror.

The coupling coefficient between the fiber and microsphere and that of the microsphere and mirror are denoted by χ_1 and χ_2 respectively.

In this section we assumed that microspheres (main sphere and its image) are isolated i.e. there are no strong perturbation effects from other nearby particles or surfaces. The symmetry associated with a mirror reflection gives the parity eigeinfunctions. Thus we obtain the even and odd eigenstates associated with blue-shifted (symmetric) and red-shifted (anti symmetric) wavelengths respectively. We can simulate the inhomogeneous space in the presence of time dependent electric field, by a volume current density distribution $(J = \varepsilon_0 \omega \Delta\varepsilon E)$ where ε_0 is the permittivity of the free space, $\Delta\varepsilon = n_s^2 - n_m^2$, E is the electric field, n_m and n_s are the refractive indices of the surrounding medium and the microsphere respectively [16]. It has been shown that the image of the electric current density parallel (perpendicular) to the flat mirror surface is symmetric (anti-symmetric) and in phase (anti-phase) with respect to the primary current density [17].

By increasing the mirror distance, the even and odd modes of mirror-microsphere system approach to the TE and TM modes of microsphere respectively. Now, the ratio of the output to the input field intensity of the even ($T_e = E_{ev} / E_i$) and odd ($T_o = E_{odd} / E_i$) modes may be calculated using the matrix method. The result is as follows:

$$T_e = \frac{r_1 - G_1 G_2 (r_2 - i\chi_2)}{1 - r_1 G_1 G_2 (r_2 - i\chi_2)} \tag{1}$$

$$T_o = \frac{r_1 - G_1 G_2 (r_2 - i\chi_2)}{1 - r_1 G_1 G_2 (r_2 + i\chi_2)}, \tag{2}$$

where G_1 and G_2 are the laser net complex gains in the upper and lower semi-spheres respectively. Due to the saturation effect and variation of the light intensity along the microsphere, G_1 and G_2 have different complex values:

$$G_i = \widetilde{G}_i e^{-\alpha L/2} e^{\frac{i\omega L n_s}{2c}} \quad i = 1,2 \tag{3}$$

where $\tilde{G}_i$ $(i=1,2)$ is the laser gain, α is the loss of the microsphere, ω is the lasing frequency, L is the circumference of the sphere, n_s is the microsphere refractive index, r_i is the transmission coefficient corresponding to the χ_i $(i=1,2)$ and c is the velocity of light. The lasing condition for the even and odd modes will be:

For even modes $$1 - r_1 G_1 G_2 (r_2 - i\chi_2) = 0 \tag{4}$$

For odd modes $$1 - r_1 G_1 G_2 (r_2 + i\chi_2) = 0 \tag{5}$$

And the oscillation frequencies of the even and odd modes (ω_e and ω_o) are also obtained by the following relations:

$$\omega_e = \frac{2m\pi c}{n_s L} + \frac{c}{n_s L} \tan^{-1} \frac{\chi_2}{r_2} \tag{6}$$

$$\omega_o = \frac{2m\pi c}{n_s L} - \frac{c}{n_s L} \tan^{-1} \frac{\chi_2}{r_2} \tag{7}$$

m is an integer about 200. For two-mode region of operation the beating frequency of the even and odd modes which can be measured easily, is related to the coupling coefficient by the following relation:

$$\chi_2 = \sin\left(\frac{\omega_B n_s L}{2c}\right) \tag{8}$$

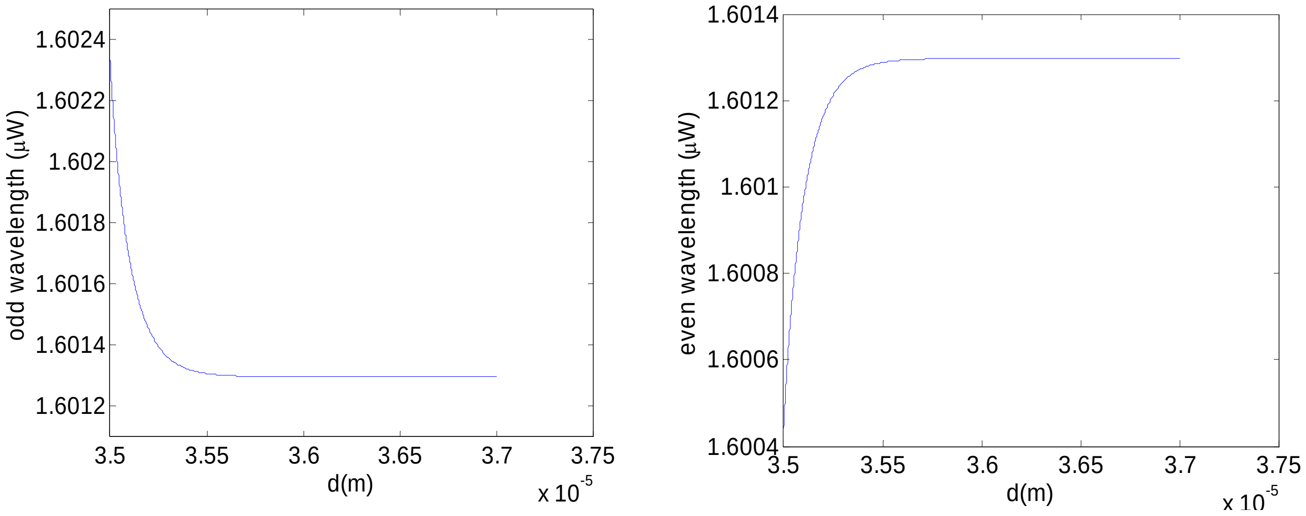
In our calculation the back scattering effects inside the microsphere are neglected [18].

The coupling coefficient χ_2, the transmission coefficient r_2 and the loss α_2 due to the microsphere-mirror coupling are related by the following relation:

$$\chi_2^{\ 2} + r_2^{\ 2} = 1 - \alpha_2 \tag{9}$$

α_2 has no significant effect on the frequency variation and will be neglected in this analysis. As it was expected the symmetric and anti symmetric modes correspond to the blue and red shifted frequencies respectively.

There are several approximation methods for calculation of the coupling coefficient [16, 19]. The method of Ref. [16] is employed in our calculations. Variation of the resonance wavelengths for odd and even modes versus the mirror-sphere distance are shown in Fig. 3a and Fig. 3b respectively.



(a) (b)

Fig. 3. Variation of the resonance wavelength for a clockwise mode versus distance *d*:
a) for odd; and b) for even mode.

When the sphere is initially located at a distance d greater than the characteristic radius R_b away from the point of contact with the mirror, the resonance frequencies are the same as that of an isolated sphere. As the sphere is brought closer to the mirror, each of the modes are splitted to red and blue shifted frequencies relative to the case where the mirror is absent. The variations of resonance frequencies are in agreement with those are obtained by the Mie scattering method [20]. The evanescent field from the microsphere surface ($R=R_o$) to the characteristic radius R_b ($R_b=\sqrt{l(l+1)}/k_0$) [8] decreases exponentially, hence by increasing the microsphere-mirror distance, the coupling coefficient decreases. High sensitivity of the coupling coefficient and frequency resonance shift of this system makes it suitable as a high sensitivity biochemical sensor even for passive microsphere resonator. The active microsphere can provide narrower line-width and hence the more sensitive biochemical sensor than those is made by passive microsphere resonator. Numerical calculations in the presence of metal mirror are in agreement with the previous published experimental results [21].

For obtaining the laser output power, the following boundary condition is used:

$$(1-\alpha_2) r_1 \widetilde{G}_1 \widetilde{G}_2 \, e^{-\alpha L} = 1 \tag{10}$$

That is the open loop gain of the system must be unity. The laser gains $\tilde{G}_i\,(i=1,2)$ are obtained by the Maxwell-Bloch equations. In this analysis, it is assumed that the mirror-microsphere coupling and tunneling losses are negligible. Hence, the cavity quality factor is limited by the fiber-microsphere coupling coefficient. The solutions of Maxwell Bloch equations are depend on the type of external mirror and generally can be obtained by the bifurcation analysis. The TE and TM modes of the microsphere are the limiting states of the even and odd modes of microsphere-mirror system when the distance between the microsphere and mirror goes to infinity. As it is well known the TE modes cannot excite the Plasmon on the surface of metal mirrors, so the attenuation of the TM mode is greater than the TE mode. Also the excited Plasmon by TM modes causes to increase the coupling coefficient between the microsphere and metallic mirror. Both of these effects cause to decrease significantly the quality factor of TM mode relative to the TE mode. Due to the decreasing of the quality factor of TM mode, the threshold power of the TM modes is significantly greater than that of TE modes, which is in agreement with the published experimental results [21]. In the absence of Rayleigh scattering and fiber-end reflections, the Clock Wise (CW) and Counter Clock Wise (CCW) modes are degenerate with the same threshold power. In the silica glass microsphere the Rayleigh scattering is not negligible and causes the frequency splitting of the CW and CCW modes [8]. The frequency splitting can be controlled by the fiber-end reflection coefficient [22]. For pump powers greater than the first threshold power (p_{th1} =82μW) and less than the second threshold value (p_{th2} =156 μW) the frequency shift of the even mode can be employed for biochemical sensor design. Due to the mode competition, for pump powers greater than the second threshold power, there are two stable and one unstable branches in the bifurcation diagram of the microshpere-metallic mirror system. For non metallic mirror system the difference between threshold pump powers is due to the difference between the Er emission cross section at the wavelength of the even and odd modes. In this analysis it is assumed that the even and odd modes have the same Er emission cross-section, hence in microsphere non metallic mirror system both even and odd modes have the same pump threshold power. Around the threshold pump power (p_{th} =64μW) the bifurcation diagram has a pitch fork configuration with two unstable and one stable branches. Below the threshold pump power, the resonance frequency can be used as a measure for a biochemical sensor. Above the threshold power, the stable mode has both the even and odd frequencies and the beating frequency between them is a suitable measure for sensor design. Due to the competition between even and odd modes and their corresponding splitting frequencies, the general bifurcation analysis is out of scope of this paper and will appear in near future. Our numerical calculations are based on the (Ro=35 μm, χ_1=0.02, Q=10, n_s=1.5, erbium concentration is denoted by N_0 , $N_0=10^{22}$ions/cm^{-3} , $S_{eff}=8\times10^{-12}$m^2 and S_{eff} is the effective mode area)

3. Sensitivity Enhancement of the active Biochemical Sensor

Our calculation is based on the homogenous broadening model and also the same method can be employed for any microsphere active medium such as Rodamine-6G doped polystyrene polymer microsphere or PbSe quantum dot silica microsphere. Oscillation frequencies are obtained by equation (6) and (7). It is easy to show that for even shifted mode the relative variation of oscillation frequency versus the variation of refractive index of surrounding medium is obtained by the following equation:

$$\frac{\Delta\omega}{\omega}=(1-\eta)\frac{\Delta n_m}{n_e}\left(-1+\frac{n_e}{2m\pi r_2}\frac{\partial\chi_2}{\partial n_e}\right) \tag{11}$$

$$\eta=\frac{\int_{cavity}|\varphi|^2 da}{\int_{total}|\varphi|^2 da}, \tag{12}$$

where η, is the fraction of power inside the microsphere and n_e is the effective refractive index of propagating mode. Equation (11) shows a refraction index variation enhancement $(n_e / 2m\pi r_2 \times \partial\chi_2 / \partial n_e)$, relative to the active microsphere biochemical sensor. The integer m is proportional to the microsphere radius of curvature ($m = n_e 2\pi R_o / \lambda$). By decreasing the microsphere size, the enhancement factor increases. Variation of $\partial\chi_2 / \partial n_e$ versus the mirror-microsphere distance is plotted in Fig. 4.

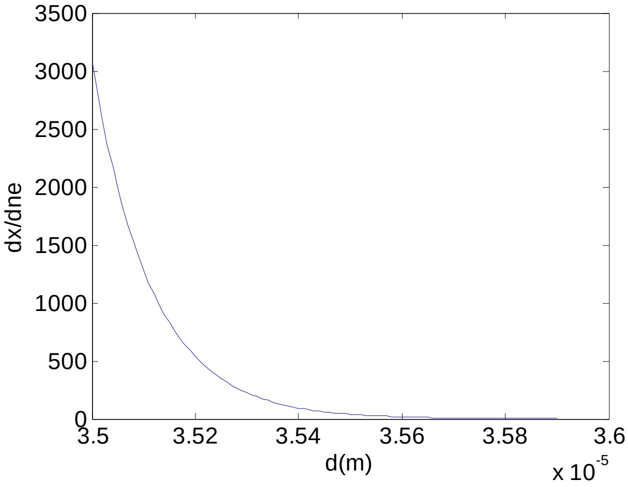


Fig. 4. Variation of $\frac{\partial\chi_2}{\partial n_e}$ versus the distance between mirror and microsphere.

Both $1/r_2$ and $\partial\chi_2 / \partial n_e$ are increasing with decreasing the microsphere-mirror distance. Variation of the enhancement factor versus the mirror- microsphere distance is shown in Fig. 5.

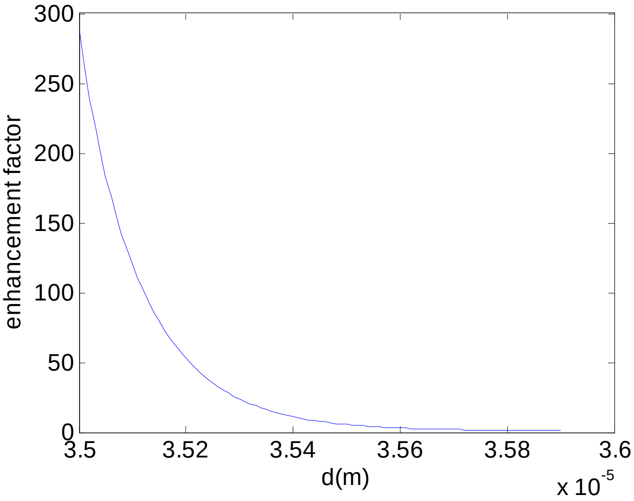


Fig. 5. Variation of the enhancement factor versus the mirror microsphere distance.

The enhancement factor causes to increase the sensitivity factor up to two orders of magnitude. From the engineering point of view, there is a lower limit for the mirror-microsphere distance and the maximum enhancement factor is not achievable. However, the engineering design of the proposed biochemical optical sensor is not in the scope of this paper. The minimum measurable frequency shift of the microsphere-mirror laser is determined by the laser line-width. The laser line-width is obtained according to the Schawlow-Townes formula for a laser oscillator [23]:

$$\Delta\omega_{osc} = \frac{N_{2th}}{\Delta N_{th}} \frac{2\pi\hbar\omega_0^3}{P_{os}\,Q^2}, \tag{13}$$

where ΔN_{th} and N_{2th} are the population inversion density and the upper laser level density at the threshold respectively. N_{2th} and ΔN_{th} are obtained from the steady state solution of the rate equations. ω_0 is the oscillation frequency of the laser oscillator and Q is the quality factor of the microsphere in the absence of active medium and in the presence of microsphere-fiber coupling. P_{os} is the laser power in the microsphere. The laser line-width is of the order of several hundred of kHz. Variation of the laser line-width and laser output power versus the laser power and pump power is obtained and presented in Fig. 6 and Fig. 7 respectively.

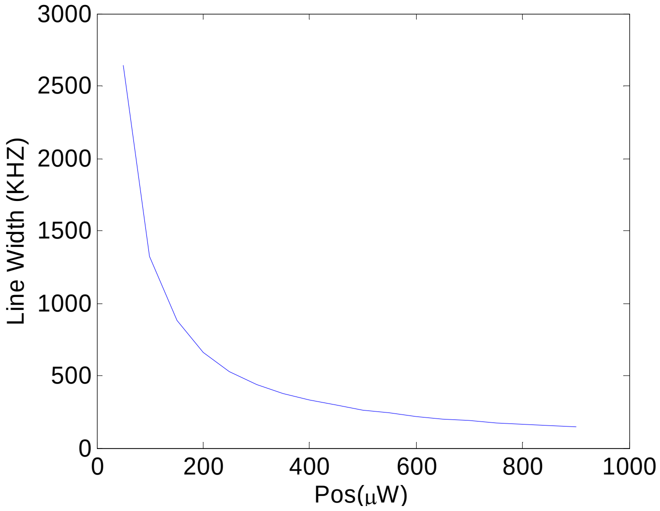


Fig. 6. Variation of the laser line width versus the laser power.

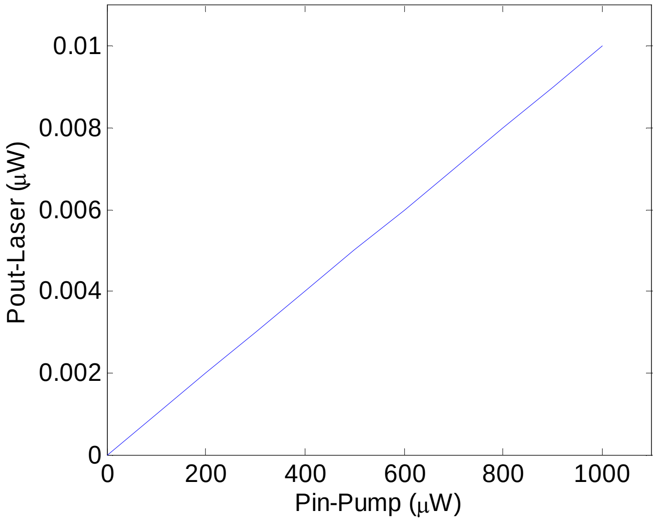


Fig. 7. Variation of the laser output power versus the pump power.

The minimum detectable change of refractive index is obtained by equations (11) and (13):

$$\frac{\Delta n_m}{n_e} = \frac{2\pi\hbar\omega_0 N_{2th}}{P_{os} Q^2 (1-\eta)(\frac{2}{mr_2}\frac{\partial\chi_2}{\partial n_e})\Delta N_{th}} \tag{14}$$

By increasing the laser power the laser line-width decreases and the minimum detectable change of refractive index also decreases, while by increasing the laser and pump powers the effects of nonlinear refractive index increase. In order to eliminate the nonlinearity effects, the biosensor can be calibrated in the presence of pump and laser powers. However, the pump and signal fluctuation could place a limit on the detectable refractive index change. For large pump power, when both even and odd modes are in stable region of operation, beating frequency between the two operating modes can be employed as a measure for the sensor design:

$$\frac{\Delta\omega_B}{\omega_B} = -2\,(1-\eta)\frac{\Delta n_m}{n_e}(\frac{n_e}{2\pi m r_2}\frac{\partial\chi_2}{\partial n_e}) \tag{15}$$

As it is expected the sensitivity is improved by a factor of two. Optomechanical oscillations of the microsphere and mirror surface produce new optical frequencies which must be eliminated through the refractive index measurement or the operating pump power is below the threshold pump power. For metal mirrors, by increasing the optical path distance between the microsphere and mirror, the laser output power increases. Hence when detection is based on fixed wavelength, the output power variation is measured due to the shift of the frequency of oscillation [24, 25]. Plasmon excitation on the metal mirror surface provides an extra sensitivity improvement. In this case, the shot noise determines the minimum power detection limit.

4. Conclusions

It is shown that the active microsphere located in the adjacent of mirror can be employed as an ultra sensitive biochemical sensor based on the laser frequency shift or power increasing. The enhancement factor relative to the active microsphere is obtained. Calculations are done for the Er^{3+} doped silica microsphere. Of course, the presented model can be employed for analysis and design of microsphere, microdisk, microtoroid or microrings doped with any active elements such as Er^{3+} ions, quantum dots, Rodamin-6G, etc. biochemical optical detectors.

Alternatively, the output laser power changes can be measured at a fixed wavelength out of the laser line-width but closed to the laser frequency as possible. In this mode of operation, also, improvement of sensitivity relative to the active microsphere biosensor is obtained.

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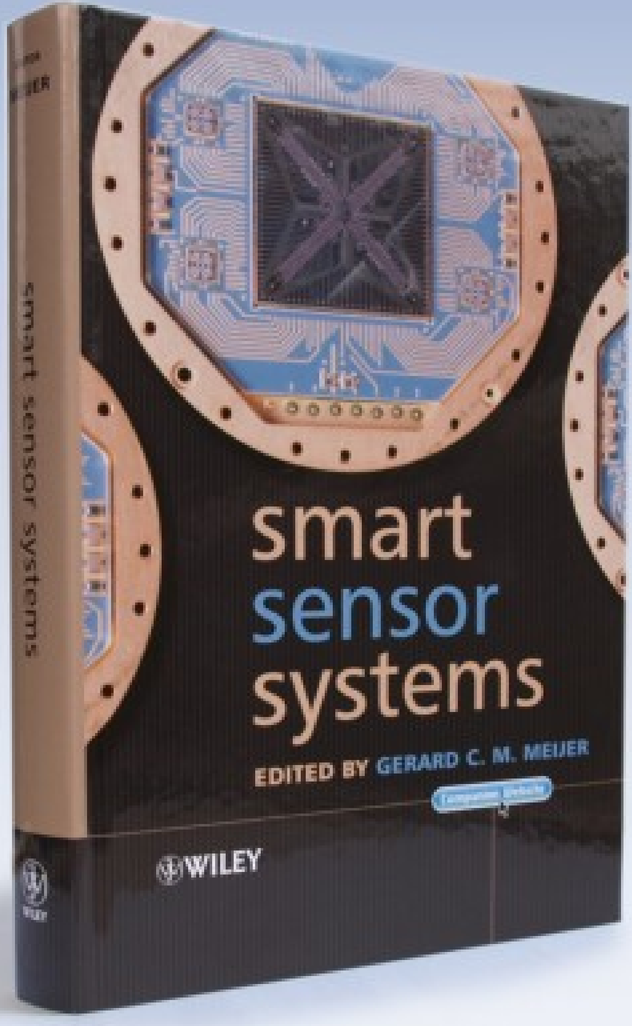
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