Interaction of a microresonator with a nanoscatterer

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Abstract. We present a theoretical analysis of the effects induced on the high-Q whispering-gallery modes of a microsphere resonator by a small metallic scatterer. We study the near-field tip-microsphere interaction varying the distance between the two spheres, in correspondence of the most perturbed modes of the microsphere. The obtained results show that this kind of interaction can be used to achieve an active control of whispering gallery resonances.

The cross section for scattering electromagnetic field by a sphere exhibits a series of sharp peaks as a function of the size parameter. These peaks indicate the presence of scattering resonances which occur when the electromagnetic field is almost trapped within the sphere [1]. A physical interpretation is that the electromagnetic wave is trapped by almost total internal reflection as it propagates around the inside surface of the sphere, and that, after circumnavigating the sphere, the wave returns to its starting point in phase. These resonances are generally referred to as morphology dependent resonances (MDRs). The degree of confinement of the mode is usually indicated by the quality (or Q-) factor that is defined like $Q = \frac{\omega}{\Delta \omega}$, where $\omega$ is the frequency of the mode and $\Delta \omega$ is the width of the corresponding narrow peak.

Optical microsphere resonators are attractive systems for both fundamental physics and for the use in optoelectronic devices [2, 3, 4]. Their very high quality factor $Q$ has attracted much attention for studies in quantum optics [5, 6], nonlinear optics [7, 8, 9, 10], optoelectronics [11, 12] and near-field optics [13, 14, 15, 16]. Their narrow linewidth has been explored in demonstrations of narrow-band passive photonic devices. By coupling MDRs with an active material, ultralow threshold lasing has been achieved [17, 18].

In particular, we are interested in studying the influence of a small metallic scatterer, placed in the near-field region of a microsphere, on the morphology dependent resonances (also known as whispering gallery modes, WGMs) supported by the microsphere. This configuration is actually relevant for several applications, because tip-sample interaction may be exploited to provide a frequency tuning of WGMs which can be of great interest for controlling the coupling of these modes with embedded or surrounding active media. Moreover WGMs can be coupled to a single emitter. The coupling can be realized by attaching the emitter to a holder that can be positioned close to the microresonator, in its evanescent field. However these nanomanipulation methods should take into account that any material that is used to hold an emitter may interact with the WGM of the microsphere, introducing additional loss mechanisms [15].

The near-field interaction of a metallic nanoscatterer with a microsphere plays a key role in apertureless scanning near-field optical microscopy (SNOM) [19]. Actually, in several applications of near-field microscopy the emitting or collecting tip is replaced by an apertureless tip placed in the near-field region of the sample. After the sample is illuminated, the tip
Figure 1. (a) Sketch of the setup. The plane wave impinges from the right along the axis joining the centers of the two spheres and the field is collected in the forward direction. (b) The dependence on the energy of the dielectric function of the nanosphere of Ag, $\epsilon = \epsilon_1 + i\epsilon_2$.

acts as a perturbing element that scatters the evanescent components nearby the sample. The influence of the tip on the light modes supported by the sample has to be understood for the correct interpretation of the measurements. It is worth noting that many near-field calculations are simplified by neglecting tip-sample coupling [20]. In many cases this is a quite good approximation [21], that however becomes questionable in presence of metallic scatterers and/or resonances as experimentally demonstrated for WGMs.

We first calculate the extinction cross section of the dielectric homogeneous microsphere through the Mie theory, to locate and classify the MDRs of the microsphere when the resonator is imperturbed [1]. Then we explore the effects produced on the most confined modes of resonance of the microsphere by approaching a metallic nanosphere at different distances. In this case, the important multiple scattering processes that occur strongly affect the optical properties of the involved scatterers [22, 23, 24, 25]. The optical properties of the system, microresonator-metallic scatterer, can be exactly calculated through the formalism of the multipole expansion of the fields in the framework of the transition matrix method: this formalism is able to take into account all the multiple scattering processes occurring among the involved spheres. Some perturbative methods have been employed to analyse two spheres systems (e.g. the iterative method of the order-of-scattering used by Fuller [26]), but they may fail to reach convergence when dealing with high-Q resonances and/or with strongly coupled spheres.

1. Transition Matrix and Multiple scattering

We assume that all the fields depend on time through the factor $\exp(-i\omega t)$, which is omitted throughout, and defining the propagation constant in vacuo $k = \omega/c$, we consider as incident field a polarized plane wave that can be expanded as

$$E_{\eta\eta}(r) = E_{0\eta} \sum_{p} \sum_{lm} J_{l,m}^{(p)}(r,k) W_{\eta lm}^{(p)},$$

where $J_{l,m}^{(p)}$ are the vector multipole fields, that are regular at the origin, $W_{\eta lm}^{(p)}$ are the (known) amplitudes of the incident field, $p$ is a parity index ($p = 1$ for magnetic multipoles and $p = 2$ for electric multipoles) and $\eta$ refers to the polarization of the incident field. The explicit expression
Figure 2. Extinction cross section $\sigma_E$ of the isolated microresonator as a function of the energy and classification of the WGMs.

for all these quantities can be found in [27].

The field scattered by the two considered spheres, which are embedded in a nonabsorptive medium, can be written as the superposition of the fields scattered by the single spheres

$$E_{S\eta} = E_0 \sum_{\alpha} \sum_{plm} H_{lm}^{(p)}(r_\alpha, k) A_{\eta\alpha lm},$$

where $H_{lm}^{(p)}$ are the multipole fields that satisfy the radiation conditions at infinity, $\alpha$ labels the single sphere (in the present case $\alpha = 1, 2$) and $r_\alpha$ is the position of the center of the $\alpha$-th sphere respect to the origin of the whole system. The amplitudes $A_{\eta\alpha lm}$ of the field scattered by each sphere are determined by the boundary conditions across the surface of each of the spheres. Their knowledge enable us to calculate all the optical properties of the scatterer and totally solves the problem of the dependent scattering from the aggregate. Actually, by imposing the boundary conditions across the surface of all the spheres, the amplitudes $A_{\eta\alpha lm}$ turn out to be the solution of the system of linear nonhomogeneous equations

$$\sum_{\alpha'} \sum_{p'l'm'} M_{\eta\alpha\alpha' l'm'} A_{\eta\alpha' l'm'} = -W_{\eta\alpha lm},$$

where

$$M_{\eta\alpha\alpha' l'm'} = R_{\alpha l}^{(p)} \delta_{\alpha\alpha'} \delta_{pp'} \delta_{ll'} \delta_{mm'} + H_{\eta\alpha \alpha' l'm'}^{(p)}$$

and $W_{\eta\alpha lm}^{(p)}$ are the amplitudes of the incident field referred to the center of the $\alpha$-th sphere.

In eq. (4) the quantities $R_{\alpha l}^{(p)}$ are the Mie coefficients of the $\alpha$-th sphere and the $H_{\eta\alpha \alpha' l'm'}^{(p)}$ the elements of the matrix that translates the origin of the $H$-multipole fields. These matrices account for the multiple scattering processes among the spheres.

2. Morphology Dependent Resonances of Microspheres

We first calculate the extinction produced in the forward direction by the isolated dielectric microsphere with refractive index $n = 2.5$ and diameter of 1 $\mu$m. Within the Mie theory for a
homogeneous sphere, we calculate the extinction cross section $\sigma_E$ of the microresonator through
the range of energies between 1.55-2.05 eV (that corresponds to $0.6 \leq \lambda \leq 0.8 \mu m$ and the size
parameter $x = kr$ varying between 3.93 and 5.24).

The extinction cross section exhibits a series of sharp peaks corresponding to the WGMs
that undergo nearly total internal reflection. Each resonance supported by the resonator is
characterized by two mode numbers, $n$, $l$, the radial and the angular one respectively, where the
last one corresponds to the multipole component that produces the mode (Fig.2). Hereafter we
will refer to the resonances modes with the symbol $\nu_{n}^{l}$.

Considering that the size of the sphere varies between $3.93 < x < 5.24$ and that its refractive
index is real, the convergence of the results is rapidly achieved to 3 significant digits with the
truncation of the multipole expansion of the fields up to $l = 9$ [28]. This choice is enough to
reach an accurate description of the fields, in particular in the range including the sharp peaks
(the MDRs) where it is more difficult to achieve the stability of the results.

3. Tip-sample interaction

When a second small metallic sphere is placed very close to the first one, so that the distances
between the centers are smaller or equal to three or four diameters (when the spheres have the
same size), the multiple scattering processes become so important that cannot be ignored and the
scattering process cannot be considered as due to independent scattering [29]. We approach to
the microsphere a small metallic sphere of Ag with radius $r = 40$nm (the size parameter being $2/25$
of the dielectric microsphere) and we illuminate the system with a linearly polarized plane wave
impinging with the wave vector along the axis joining the centers of the two spheres (Fig. 1a). For
the dependence of the dielectric function of silver on the energy of the incident field we refer to
the experimental values provided by Johnson and Christy [30] (see Fig. 1b). The small metallic
sphere is placed in the evanescent field of the WGMs of the microresonator and we calculate
the extinction cross section in the forward direction of the whole system. Furthermore, we
underline that we do not need to introduce any extra-term to the expansion of the fields to take
into account the contribution of the evanescent components of the field, because the multipole
expansion gives a complete description of the electromagnetic fields without introducing any
approximation, except the natural truncation of the expansion. The evanescent components of
the scattered field are indeed intrinsically considered.

Introducing the metallic nanoscatteerer, although the overall size parameter of the system
(dielectric microsphere and metallic nanosphere) does not change much compared to the size
of the isolated dielectric microsphere ($4.26 < x_{system} < 5.86$), several conditions make more
difficult to achieve an easy convergence of the results: (i) the appreciable difference in size of the
two spheres ($r/R = 2/25$), (ii) the small mutual distance between the surfaces of the spheres that
induces multiple scattering processes occuring with noticeable strength, (iii) the occurrence of
resonances with very narrow peaks, (iv) the high value of the immaginary part of the refractive
index of the small sphere (i.e., dealing with a metallic particle). In these conditions, to calculate
the optical properties of the two spheres system we need to extend the multipole expansion up
to $l=15$ to ensure the numerical stability of the results and reach an accuracy of 3 significant
digits [31].

4. Results

We place the tip at $5 < d < 40$nm where $d$ is the distance between the surfaces of the two
scatteres. To analyse the effects of the coupling of the spheres, we concentrate on the
most perturbed modes of the isolated dielectric sphere: $\nu_{4}^{1}, \nu_{3}^{2}, \nu_{1}^{2}$. These correspond to
the most confined resonances of the microresonator, i.e., the resonances with the highest Q-factor:
respectively $1.66 \cdot 10^4$, $4.26 \cdot 10^3$, $5.57 \cdot 10^3$.
The introduced perturbation produces a degradation of the Q-factor as a result of scattering
losses as well as a shift of frequency in agreement with the experimental observations [14, 15]. In Table 1 we present the values of the ratio \( Q/Q_0 \) corresponding to different distances of the nanoscatelar from the microresonator (with \( Q_0 \) we define the unperturbed Q-factor of the isolated dielectric sphere). As expected, the Q-factor reduction is more pronounced the bigger the initial \( Q_0 \) is, and the degradation of the peak decreases with the increase of the distance \( d \). Actually, the values of \( Q/Q_0 \) corresponding to the most confined modes, \( \nu_4 \) and \( \nu_8 \), are more perturbed than the \( \nu_2 \), although this one corresponds to a higher value of energy than the \( \nu_8 \). Indeed, the degree of perturbation is mainly linked to the value of the Q-factor than to the energy of the resonance. In Fig.3 the energy shift undergone by the three considered resonances are shown plotting \( E_0(\nu^n_l) - E(\nu^n_l, d) \) versus the distance (with \( E_0 \) we refer to the energy of the unperturbed resonance). When the tip approaches the microresonator the peaks shift towards lower energies. This effect becomes more evident the shorter the distance between the two spheres is.

We investigated theoretically the problem of a small metal sphere in the near-field of a high-Q dielectric resonator that supports various whispering gallery modes. The obtained results show that the higher the Q-factor of the resonance the more sensitive the resonance is to external perturbations [15]. This means that the effects we observed should be even more relevant in microresonators with larger size parameter. However, thanks to the small size parameter of the microresonator, we have been able to obtain relevant informations on the tip-microsphere
interaction with a reduced computational effort. The method we presented can be particularly useful for studying a number of interaction processes in presence of resonances. Furthermore, our theoretical analysis of the problem of tip-sample interaction has direct applications in apertureless near-field microscopy when the sample supports high-Q resonances. In particular, our calculations could be relevant for the experimental research that is presently carried on in many laboratories that study biochemical reactions occurring on a glass sphere placed at the end of a fiber. The possibility of probing the glass sphere surface with a scanning near-field optical microscope (SNOM) makes our results even more useful.

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