

Rayleigh scattering of whispering gallery modes of microspheres due to a single dipole scatterer

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An exact solution for the problem of interaction between whispering gallery modes of spherical microresonators and a single dipole is presented. It is predicted that experimentally observed spectral doublets associated with this interaction are a part of a triplet whose third component has yet to be observed. It is also shown that the interaction between the dipole and nonresonant whispering gallery modes results in a renormalization of the dipole's polarizability and experimental manifestations of this effect are discussed.

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I. INTRODUCTION

In this Rapid Communication we present a rigorous *ab initio* theory of interaction between whispering gallery modes (WGMs) of spherical resonators and a single small nonresonant dielectric scatterer. Results of paper [1], where this phenomenon was studied experimentally, indicated that the single-defect scattering reproduces effects typical for scattering of WGMs due to technological subwavelength imperfections of the resonators. Accordingly, the results of the presented theory allow at least qualitative discussion of this more general situation as well. The relevance of the single-defect problem for the case of multiple scatterers can be justified by noting that this scattering is usually weak and can be treated in a single-scattering approximation when each of multiple imperfections behaves as an independent defect. The developed theory also lays foundation for solving more complex problems such as interaction between WGMs and defects with internal resonances such as quantum dots, which has recently attracted attention in the context of quantum information [2].

The scattering of WGMs significantly affects optical properties of microresonators and has, therefore, attracted a great deal of attention [3–7]. It is believed that one of its manifestations is splitting of a spectral peak corresponding to an individual WGM into a spectral doublet [3–6]. Results of Ref. [1], where double peak features in the spectra of microspheres were studied under conditions of controlled scattering due to a single intentionally introduced defect directly confirmed this assertion. The currently accepted explanation of this phenomenon is based on the *ad hoc* introduced concept of defect-induced coupling between two degenerate counterpropagating clockwise (cw) and counterclockwise (ccw) WGMs. According to this concept the coupling removes the degeneracy of the modes and splits respective frequencies [3–7]. This approach is very appealing due to its conceptual simplicity and has become paradigmatic in optics of microresonators. However, results of rigorous calculations presented here show that it is inadequate and does not reproduce even qualitatively many of essential features of the phenomenon under consideration. On the very basic level the main shortcoming of the “backscattering paradigm” consists in treating essentially quasi-two-dimensional WGMs in microspheres in the one-dimensional approximation. It also ignores the vector nature of electromagnetic field, which manifests itself in this problem in the form of dependence of the

strength of WGM-defect interaction on orientation of the dipole moment of the defect.

II. THEORETICAL FORMALISM

Our theory is based on the observation that if a scatterer, as it is usually assumed (see, e.g., Ref. [1]), can be treated as a dipole, its shape is irrelevant and can be taken to be spherical. The resulting situation of two electromagnetically coupled dielectric spheres of radii R_0 and R_d ($R_d \ll R_0$), characterized by refractive indexes n and n_d , respectively, and positioned at a distance d from each other (see Fig. 1) submits to rigorous *ab initio* treatment based on the standard multisphere Mie formalism [8,9]. More specifically, we will consider interaction between TM-polarized fundamental mode (FM) of a spherical resonator, characterized by polar number L , radial number $s=1$, and field concentrated in one the sphere's equatorial planes. Electric field of such a mode shows no oscillations along radial and polar coordinates and is characterized by a smallest mode volume. We will also assume that the defect is positioned near the surface of the sphere in the plane of the mode, where the interaction is strongest, but the formalism developed applies to arbitrarily positioned scatterers.

FMs are habitually described in a coordinate system where the polar axis is perpendicular to the plane of the mode ($X'Y'Z'$ system in Fig. 1). In this system a FM is characterized by a single azimuthal number $|m|=L$. However, the defect-induced loss of the rotational symmetry with respect to axis Z' makes it more convenient to switch to a different coordinate system with the Z axis passing through the centers of the spheres (XYZ system in Fig. 1). The field distribution corresponding to the FM shown in this figure

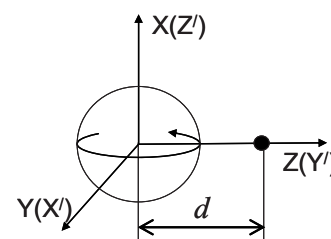


FIG. 1. The coordinate systems used in calculations and the original FM interacting with the defect (blackened small sphere).

cannot, of course, be described in the new coordinates by a single mode [10]. Instead, one needs a linear combination of WGMs with given polar number L and all azimuthal numbers $|m| \leq L$, which can be written down as

$$\mathbf{E}_{\text{FM}} = \sum_{m=-L}^L \eta_{L,m} \mathbf{N}_{L,m}^{(1)}(\mathbf{r}), \quad (1)$$

where $\mathbf{N}_{L,m}^{(1)}(\mathbf{r})$ is a vector spherical harmonic (VSH) of TM polarization with radial dependence given by spherical Bessel functions, as defined in Ref. [9]. Coefficients $\eta_{L,m}$ can be found from transformation properties of the spherical harmonics [9]; their explicit form is provided in the supplementary materials of this Rapid Communication [11].

The goal of the theory is to find the spectrum and spatial distribution of the electromagnetic field induced in and around the structure under consideration by an incident wave (imitating a mode of a tapered fiber) which, in the absence of the defect, would have excited a field distribution given by Eq. (1). Following the standard multisphere Mie theory [9] we present the induced field exterior to the sphere, \mathbf{E}_s , as a linear combination of VSH with different polar, l , and azimuthal, m , numbers:

$$\mathbf{E}_s = \sum_{i=1}^2 \sum_{l=1}^{\infty} \sum_{m=-l}^l a_{l,m}^{(i)} \mathbf{N}_{l,m}^{(3)}(\mathbf{r} - \mathbf{r}_i), \quad (2)$$

where index i enumerates the spheres ($i=2$ refers to the defect), \mathbf{r}_i is a position vector of the center of i th sphere, and the radial dependence of the VSH is given by the outgoing spherical Hankel function. Strictly speaking this expansion must also contain VSHs of TE polarization, but cross-polarization coupling is weak in the situation under consideration and can be incorporated perturbatively. Using a similar presentation for the internal fields, subject to Maxwell boundary conditions, one can derive a system of linear equations for the expansion coefficients $a_{l,m}^{(i)}$:

$$a_{l,m}^{(i)} = \alpha_l^{(i)} \left[\eta_{l,m} \delta_{L,l} \delta_{i,1} + \sum_{j \neq i} \sum_{l'} a_{l',m}^{(j)} A_{l',m}^{l,m}(\mathbf{r}_j - \mathbf{r}_i) \right], \quad (3)$$

where $\alpha_l^{(i)}(x)$ is the single sphere Mie scattering parameter for TM polarization dependent on dimensionless frequency parameter $x = R_0 \omega / c$. The real and imaginary parts of the complex zeroes of $[\alpha_l^{(1)}(x)]^{-1}$ determine, accordingly, frequencies, and lifetimes of the respective WGM, while $\alpha_l^{(2)}$ does not have any poles due to the small size of the defect. The explicit expression for $\alpha_l(x)$ is given in [11]. The electrodynamic interaction between spheres is described by translation coefficients $A_{l',m}^{l,m}$, which enter the theory when one applies the vector addition theorem [9] to VSH centered at different spheres; convenient formulas for these coefficients can be found in Refs. [8,9]. Translation coefficients are diagonal in m in Eq. (3) which is consequence of the axial symmetry of the resonator-defect structure with respect to the polar axis of the XYZ coordinate system. If we stayed in the $X'Y'Z'$ coordinate system the nondiagonal elements of the translation coefficients would have coupled modes with all azimuthal numbers and not just counterpropagating modes with $m=L$ and $m=-L$ as assumed in the backscatter-

ing paradigm. In the XYZ system the fields are presented as combinations of independent m components, which represent *eigenmodes* of the resonator-defect system. Consequently, the result of interaction between the resonator and the defect is most conveniently discussed in terms of these eigenmodes and their respective eigenfrequencies.

The dipole approximation for the defect is introduced by setting all $a_{l,m}^{(2)} = 0$ for $l \geq 2$, which reflects the fact that the field of a dipole is completely described by TM polarized VSHs with $l=1$. The remaining (still infinite) system of equations for the field coefficients $a_{l,m}^{(1)}$ can be solved *exactly* [11]. The solution shows that only those $a_{l,m}^{(1)}$ for which $m \leq 1$ are affected by interaction with the dipole so that all coefficients with $|m| > 1$ reduce to the single sphere expression $a_{l,m}^{(1)} = \alpha_l^{(1)} \eta_{L,m} \delta_{l,L}$. This result simply reflects the absence of multipole components with $m > 1$ in the dipole field of the defect. The contribution of these coefficients to the induced field [Eq. (2)] is responsible for the resonance at the unperturbed single sphere frequency, $x_L^{(0)}$ with width, $\gamma_L^{(0)}$, also unaffected by the defect.

The coefficients $a_{l,m}$ with $m \leq 1$ and $l=L$ are found as

$$a_{L,m} = \frac{\eta_{L,m} (1 + \alpha_1^{(2)} \sigma_L)}{[\alpha_L^{(1)}]^{-1} (1 + \alpha_1^{(2)} \sigma_L) + (-1)^L \alpha_1^{(2)} A_{1,m}^{L,m} A_{L,m}^{1,m}}, \quad (4)$$

where the term $\sigma_L = \sum_{\nu \neq L} (-1)^\nu \alpha_\nu^{(1)} A_{1,m}^{\nu,m} A_{L,m}^{1,m}$ takes into account the interaction between the defect and all modes with $l \neq L$. The poles of these coefficients are shifted with respect to single sphere positions, so that the contribution of these coefficients to the induced field gives rise to new resonances.

III. RESULTS AND DISCUSSION

Assuming that the shift is small compared to the single sphere frequency and that σ_L does not contain any resonant terms in the frequency range around $x_L^{(0)}$, we can find an approximate expression for new resonance frequencies, $x_{L,m}$, and their widths $\gamma_{L,m}$ which we present in the following form: $x_{L,m} = x_L^{(0)} + \delta x_{L,m}$ and $\gamma_{L,m} = \gamma_L^{(0)} + \delta \gamma_{L,m}$, where $\delta x_{L,m}$ and $\delta \gamma_{L,m}$ are

$$\delta x_{L,m} = -\gamma_L^{(0)} p [x_L^{(0)}]^2 \frac{f_{L,m}}{(2L+1)R_0^2 d}, \quad (5)$$

$$\delta \gamma_{L,m} = \frac{2}{3} \gamma_L^{(0)} p^2 [x_L^{(0)}]^5 \frac{f_{L,m}}{(2L+1)R_0^5 d}, \quad (6)$$

and all frequency-dependent quantities are calculated at $x = x_L^{(0)}$. Parameter p introduced in Eqs. (5) and (6) is defined as

$$p = \frac{p_0}{1 + \frac{2p_0}{3R_0^3} \text{Im}[\sigma_L]}, \quad (7)$$

where $p_0 = (n_d^2 - 1) / (n_d^2 + 2) R_d^3$ is the standard dipole polarizability of a dielectric sphere. This parameter has the physical meaning of effective position-dependent polarizability renormalized by interaction between the dipole and nonresonant

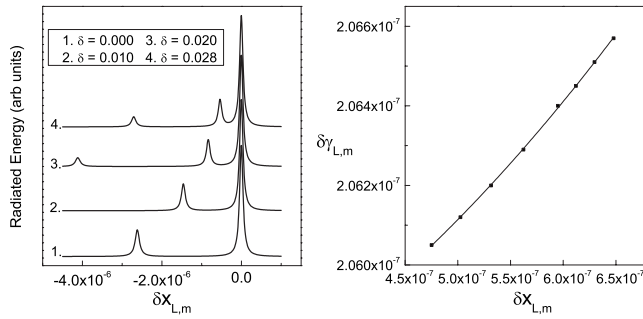


FIG. 2. Scattered power for different distance parameter $\delta = (d - R_0 - R_d)/R_0$ (left); relative broadening versus frequency shift of the $|m|=1$ resonance revealed through varying distance d (right). The line shows a fit with a quadratic polynomial.

($l \neq L$) multipole fields entering Eq. (7) via factor σ_L . Finally, function $f_{L,m}(x_L^{(0)}d/R_0)$ describes the dependence of the new frequencies upon azimuthal number m and distance d . It is positive for all values of its argument and is invariant with respect to replacement of m to $-m$; its explicit form is given in the supplemental materials [11]. Since this function is different for $m=0$ and $m=\pm 1$, Eqs. (5) and (6) describe two defect-induced resonances, both redshifted from $x_L^{(0)}$, which are produced by different components of the FM. The $m=0$ component interacts more strongly with the dipole, resulting in both a larger frequency shift and radiative broadening as compared to the resonance produced by the interaction with $|m|=1$ components.

Even though the incident field contains only the WGM with $l=L$, the intermode coupling results in excitation of nonresonant modes characterized by coefficients $a_{l,m}^{(1)}$ with $l \neq L$ and $|m| \leq 1$. If there are no single sphere resonances with $l \neq L$ in the frequency range of interest, these coefficients resonate at the same frequencies as coefficients with $l=L$, but their contribution to the total field is small. Explicit expression for these coefficients as well as for coefficients $a_{l,m}^{(2)}$ describing defect contribution to the field is given in [11].

Thus, instead of a traditional doublet, our theory predicts the existence of a triplet of peaks whose physical meaning can be explained as follows. All eigenmodes of the defect-resonator system are divided in three groups: those which are not affected by the defect ($|m| > 1$), an eigenmode with dipole moment of the defect oriented along the Z axis ($|m|=0$), and two degenerate eigenmodes with the dipole moment in the XY plane of the coordinate system. The splitting between $m=0$ and $|m|=1$ modes reflects a difference between interaction strength for two different orientations of the dipole. Since the initial FM of the resonator is comprised of WGMs with all values of m , it demonstrates resonant response at all three frequencies. This description of the sphere-defect interaction in terms of true eigenmodes of the resonator-defect structure gives a physically correct picture of the scattering of WGMs which replaces the backscattering paradigm.

These resonances are shown in Fig. 2, where we plot the spectrum of the electromagnetic power radiated by the resonator-dipole system obtained by exact integration of the

Poynting vector of the total scattered field over a closed surface in the far field region. The calculations were carried out for $L=39$ and take into account all expansion coefficients $a_l^{(2)}$ and $a_l^{(1)}$ with $l < l_{max}$, where l_{max} was chosen to ensure convergence of the calculations. A special procedure for improving numerical convergence of σ is described in [11]. As expected the peak corresponding to $m=0$ is farther away from the single sphere resonance and is weaker than the peak associated with $m=\pm 1$ so that we assume that the experimentally observed “doublet” corresponds to the single sphere and $m=\pm 1$ resonances. Indeed, experimental results of Ref. [1] showed that only the lower frequency peak shifts with the change in the defect’s distance d while the higher frequency part of the doublet does not depend on d . It is interesting to note that the condition for resolving these resonances $\delta\gamma_{L,m}/\delta x_{L,m} \propto (x_L^{(0)}R_d/R_0)^3 < 1$ coincides with the condition of applicability of the dipole approximation for the defect.

The position dependent renormalization of the defect’s polarizability described by Eq. (7) might have contributed to another experimental finding of Ref. [1]. Varying defect distance d , Mazzei *et al.* studied the relation between $\delta x_{L,m}$ and $\delta\gamma_{L,m}$. Instead of the expected linear proportionality this relation was found to be approximately quadratic. Since $\delta x_{L,m} \propto p$ while $\delta\gamma_{L,m} \propto p^2$, it was suggested in Ref. [1] that this result can be explained by position dependent renormalization of the defect polarizability p due to deviations from the dipole approximation. The theory presented here shows that the renormalization of the polarizability can also be a result of interaction between the dipole and nonresonant WGMs. In order to elucidate this possibility we determined $\delta x_{L,m}$ and $\delta\gamma_{L,m}$ for the $|m|=1$ resonance from spectra obtained for different distances d , and found the quadratic dependence shown in the left panel of Fig. 2.

Having found the scattering coefficients we can also compute expansion coefficients of the internal field and study the distribution of its intensity along the sphere. The results of the calculations for the plane of the initial FM are shown in Fig. 3 for the $|m|=1$ defect-induced and single-sphere resonances. These plots demonstrate oscillatory distribution of the field’s intensity with $2L$ oscillations. The field distribution at the $|m|=1$ resonance is characterized by a strong two-pronged enhancement of the field at the position of the defect. This intensity distribution is explained by the fact that the total field at this frequency is mainly comprised of the multipole components with $|m|=1$, which in the YZ plane are strongly peaked near the poles at $\theta=0$ and $\theta=\pi$ for both $\phi=\pi/2$ and $\phi=3\pi/2$. Each component undergoes $L-|m|+1=L$ oscillations in the half plane of $0 \leq \theta \leq \pi$ for a total of $2L$ oscillations. The field distribution at the single sphere resonance results from the exclusion of $|m|=1$ and $m=0$ contributions from the originally uniform intensity distribution of the unperturbed FM. This field distribution accounts for the backpropagating waves observed in Ref. [6] without recourse to the backscattering hypothesis.

IV. CONCLUSION

The theory of interaction between WGMs of microspheres and a subwavelength scatterer presented in this Rapid Com-

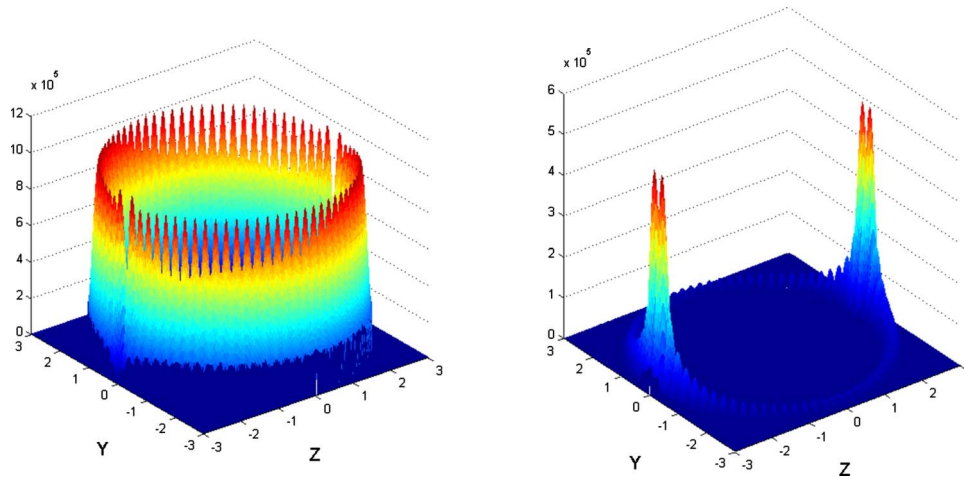


FIG. 3. (Color online) Internal field intensity of the microsphere in the YZ plane at the frequency of the standard Mie resonance (left) and the defect-induced resonance (right).

munication offers a physical picture of this phenomenon based on consideration of eigenmodes of the defect-resonator structure. The theory predicts that this interaction results in formation of a triplet of resonances in stark contrast with backscattering paradigm, which predicts only a doublet of peaks. The yet unobserved third component of the triplet can be found in an experiment similar to that of Ref. [1], where one should look for an additional peak whose frequency would depend on the position of the scatterer. It is also found that interaction of the defect with higher-order WGMs results in the position-dependent renormalization of its polarizability. Manifestations of this effect might have been observed in Ref. [1]. In addition, the developed theory allows consider-

ing other effects such as scattering-induced polarization conversion, which will be discussed elsewhere. Finally, it is clear that the presented physical picture and main qualitative conclusions of the Rapid Communication are robust with respect to small deviations of the resonator from ideal sphericity.

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