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Propagation of the fundamental whispering gallery modes in a linear chain of microspheres

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Abstract Multi-sphere Mie theory is used to analyze the resonance response and the spatial distribution of the electromagnetic field in a system of a linear chain of microspheres. We assume that the system is coupled to a monochromatic source that would have excited a fundamental whispering gallery mode in a single sphere, and study the modification of the resonance frequencies and the spatial distribution of the field induced by optical coupling between spheres. We find that the coupled-mode approach does not give an adequate description of this situation, and that the excitations of the chain cannot be presented as linear combinations of the single-sphere fundamental modes. In the case of chains with an odd number of microspheres, there exists a single collective mode with frequency equal to that of the single-sphere resonance, which reproduces the field pattern of a single-sphere fundamental mode without distortion in every odd-numbered sphere of the chain.

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1 Introduction

Optical microresonators attract a great interest due to their potential for a variety of applications [1] and recent technological advances allowing for very accurate manufacturing of the respective structures [2]. Individual resonators of various shapes such as microspheres [3], microdisks [4], microtoroids [5] etc. have been carefully studied and shown to have practically achievable Q-factors (Q) as large as 10^{11}–10^{13}. Of particular interest are structures in which several microresonators are optically coupled by evanescent fields of their individual modes. The simplest such configuration is a double-resonator photonic molecule suggested in [6] and studied both experimentally and theoretically in many papers [7–9]. More complex configurations such as triangular, square, hexagonal, or circular structures were also studied [2, 10]. A number of new devices such as, for instance, coupled-resonator optical waveguides (CROWs) were proposed based on coupling of a large number of high-Q resonators [11].

In the case of microspheres, resonances are whispering gallery modes (WGMs) characterized by angular quantum number l, azimuthal number m, radial number s, and polarization, TE or TM. In order to achieve high values of Q-factor one needs to excite WGMs with large values of l, which are characterized by a high degree of degeneracy. Modes with the same orbital and radial numbers, l and s, but different azimuthal numbers m have the same (complex-valued) frequency, but different spatial distributions. Not all of the degenerate modes are equally beneficial for applications (even though they all have the same Q-factor), because they are characterized by different values of the mode volume and the surface field enhancement. Of greatest interest are so-called 'fundamental' modes, which are characterized by $s = 1$ and $|m| = l$. The field of these modes is tightly concentrated at the surface of the sphere in the vicinity of the equatorial plane, resulting in the smallest mode volume and greatest surface field enhancement. It is assumed that these modes can be excited, for instance, by coupling to a tapered fiber [12].

When microspheres are arranged in coupled structures one would like to achieve a field distribution resulting from coupling between the fundamental modes of individual spheres. In the spirit of a popular coupled-mode approach [11] such a distribution is usually described as a linear combination of phase-matched ($m = l$ in one sphere and $m = -l$ in the other) modes whose coupling is characterized by an overlap integral of the respective modal functions [13, 14]. Similar results are obtained in the first order of the perturbation theory applied to a system of two or more resonators [14].

We show in this paper that field configurations in the form of symmetric and anti-symmetric combinations of single-sphere modes are not consistent with the symmetry of the system if the deviations of the spheres from the ideal shape are negligible. More accurately, we demonstrate that if one excites a fundamental mode in each of uncoupled ideal individual spheres and brings them together for optical coupling, the ensuing violation of the complete spherical symmetry of the system will result in a field configuration containing a linear combination of all initially degenerate modes with $|m| \leq l$. The optical coupling removes the degeneracy of these modes, producing a complicated optical response with multiple peaks.

This situation cannot be described by the non-degenerate perturbation approach used in [14] or, equivalently, by the mode-coupling theories even in the case of weak coupling because of the degeneracy of the unperturbed system. In the language of the perturbation theory, the modes of individual spheres do not form correct zero-order eigenfunctions, and one needs to find a correct basis, which would diagonalize the perturbation matrix. We will show here that an approach based on a multi-sphere Mie theory allows one not only to solve the problem of optical coupling of linear chains of microspheres numerically with a higher degree of accuracy, but it also provides the most natural way of developing an approximate description of the problem free of difficulties associated with direct perturbative assault of the Maxwell equations.

More specifically, we will describe in this paper the optical response and the field distribution in a linear chain of coupled spheres under the excitation conditions that would have generated fundamental modes in an isolated sphere, using a combination of numerical and approximate analytical calculations. The general results will be applied to particular cases of $N = 2, 3, 4$, and 5 spheres.

2 Fundamental modes and coordinate systems

The system considered in the paper consists of a linear chain of N dielectric spheres of radius R and refractive index n positioned at a distance $d \geq 2R$ between their centers (an example with the number of spheres $N = 2$ is shown in Fig. 1). We assume that there is a monochromatic incident field, $\mathbf{E}_{\text{inc}}$, of frequency ω, exciting certain WGM resonances in the system, which are described by scattered, $\mathbf{E}_{\text{sc}}$, and internal, $\mathbf{E}_{\text{in}}$, fields. Our task is to find the resonance frequencies and the total field distribution in and around the spheres, which we solve with the help of the standard multi-sphere Mie theory [9, 15, 16]. In this approach one divides the electromagnetic field in the region of space occupied by spheres into incident, internal, and scattered fields. Considering an ith sphere whose center is located at point $\mathbf{r}_i$, one presents the field incident at this sphere, $E^{(i)}_{\text{inc}}$, and the respective internal field, $E^{(i)}_{\text{in}}$, as linear combinations of single-sphere vector spherical harmonics (VSH) $\mathbf{M}_{m,l}$ (TE polarization) and $\mathbf{N}_{m,l}$ (TM polarization) centered at the chosen sphere:

$$\mathbf{E}^{(\mathbf{i})}_{\text{inc}} = \sum_{l,m} \left[\zeta^{(i)}_{l,m} \mathbf{N}_{m,l}(\mathbf{r} - \mathbf{r}_i) + \eta^{(i)}_{l,m} \mathbf{M}_{m,l}(\mathbf{r} - \mathbf{r}_i) \right], \tag{1}$$

$$\mathbf{E}^{(\mathbf{i})}_{\text{in}} = \sum_{l,m} \left[c^{(i)}_{l,m} \mathbf{N}_{m,l}(\mathbf{r} - \mathbf{r}_i) + d^{(i)}_{l,m} \mathbf{M}_{m,l}(\mathbf{r} - \mathbf{r}_i) \right]. \tag{2}$$

The scattered field is expressed as a sum of the fields scattered by each sphere:

$$\mathbf{E}_{\mathbf{s}} = \sum_{i=1}^{N} \sum_{l,m} \left[a^{(i)}_{l,m} \mathbf{N}_{m,l}(\mathbf{r} - \mathbf{r}_i) + b^{(i)}_{l,m} \mathbf{M}_{m,l}(\mathbf{r} - \mathbf{r}_i) \right]. \tag{3}$$

Using Maxwell boundary conditions and the addition theorem for the vector spherical harmonics [17, 18], one can derive a system of equations relating the scattering coefficients $a^{(i)}_{l,m}$ and $b^{(i)}_{l,m}$ to the coefficients of the incident field

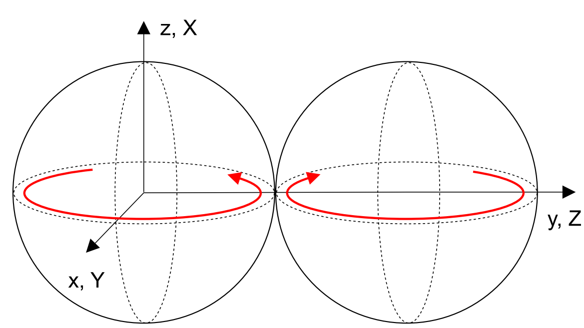


Fig. 1 Configuration of a two-sphere system and possible coordinate systems. *Arrowed lines* show schematically two single-sphere fundamental modes

$\zeta^{(i)}_{l,m}$ and $\eta^{(i)}_{l,m}$:

$$a^{(i)}_{l,m} = \alpha^{(N)}_l(x)\Big\{\zeta^{(i)}_{l,m} + \sum_{j\neq i}\sum_{l',m'}\big[a^{(j)}_{l',m'}A^{l',m'}_{l,m}(x,\mathbf{r}_j-\mathbf{r}_i) + b^{(j)}_{l',m'}B^{l',m'}_{l,m}(x,\mathbf{r}_j-\mathbf{r}_i)\big]\Big\}, \tag{4}$$

$$b^{(i)}_{l,m} = \alpha^{(M)}_l(x)\Big\{\eta^{(i)}_{l,m} + \sum_{j\neq i}\sum_{l',m'}\big[b^{(j)}_{l',m'}A^{l',m'}_{l,m}(x,\mathbf{r}_j-\mathbf{r}_i) + a^{(j)}_{l',m'}B^{l',m'}_{l,m}(x,\mathbf{r}_j-\mathbf{r}_i)\big]\Big\}, \tag{5}$$

where $\alpha^{(N)}_l$ and $\alpha^{(M)}_l$ are single-sphere Mie scattering coefficients for TM and TE polarizations, respectively, which have poles at the specific values of the dimensionless frequency parameter x, defined as $x = nR\omega/c$. Real and imaginary parts of these poles determine the frequency and the spectral width of the WGM resonances of single spheres. Explicit expressions for the scattering coefficients as well as for the translational coefficients $A^{l',m'}_{l,m}(\mathbf{r}_j - \mathbf{r}_i)$ and $B^{l',m'}_{l,m}(\mathbf{r}_j - \mathbf{r}_i)$, which describe optical coupling between the spheres via modes of the same or different polarizations, respectively, can be found, for instance, in [9, 15, 16]. One, however, needs to be careful because different authors use different normalization for VSH, which also affects the definition of the translation coefficients. We will use here the formulation presented in [15].

Our goal is to study effects of the optical coupling on fundamental modes of single spheres. One has to realize, however, that the classification of WGMs as fundamental is linked to a particular choice of a coordinate system: the mode with $|m| = l$ is associated with an equatorial plane perpendicular to the polar axis z. When dealing with a single-sphere problem, one can designate any plane passing through the sphere's center as an equator and have a fundamental mode assigned to it by choosing a polar axis of the spherical coordinate system in the appropriate direction. The situation changes, however, when one wants to optically couple fundamental modes of two or more spheres. In this case, in order to achieve maximum coupling, one would like to have field distributions, which, in the absence of coupling, would have been concentrated in the vicinity of the plane containing the centers of the spheres. In order to classify these fields as fundamental modes, one has to designate this plane as equatorial by choosing a polar axis of the coordinate system to be perpendicular to the line connecting the centers of the spheres. This coordinate system is labeled by lower case letters in Fig. 1.

Using this coordinate system one can simulate coupled fundamental modes of a definite polarization, for instance TE, and with a given angular momentum L by choosing expansion coefficients for the incident field, $\mathbf{E}_{\rm inc}$, coupled to the first sphere as

$$\zeta^{(1,2)}_{l,m} = 0, \qquad \eta^{(i)}_{l,m} = \delta_{lL}\delta_{mL}\delta_{i1}, \tag{6}$$

and assuming that its frequency is in the vicinity of the resonance with the lowest radial number, $s = 1$. The incident field chosen in the form of (6) excites in a single sphere a required high-Q fundamental mode with $l = L$ and $m = L$, so that this choice enables us to study effects of optical coupling on this particular mode. However, this choice of a coordinate system is not consistent with the symmetry of the linear chain and, as a result, the azimuthal number m does not conserve in this system. The formal manifestation of this fact is the presence of non-diagonal in m elements in the translation coefficients, which mixes single-sphere modes with different azimuthal numbers. As a result, the configuration of the fields in the coupled spheres cannot be presented as a combination of the modes with $|m| = l$, contrary to the assumption of the coupled-mode theories.

While one can solve (4) and (5) using the xyz coordinate system with the coefficients of the incident field given by (6), the non-diagonal in m nature of the translation coefficients makes calculations too complicated and masks the physical meaning of the effects under discussion. It is more convenient to use a different coordinate system with polar axis along the line of symmetry of the system (XYZ system in Fig. 1), in which the translation coefficients are diagonal in m and the normal modes of the coupled system can again be classified according to the azimuthal number [9, 19]. However, the field distribution corresponding to the fundamental mode of the xyz coordinate system cannot be characterized by a VSH with a single m in the XYZ system. In order to describe the same field distribution using new coordinates one has to use transformation properties of VSH [15]. As a result the incident field given by (6) in xyz coordinates is described now by coefficients

$$\zeta^{(i)}_{l,m} = 0, \qquad \eta^{(i)}_{l,m} = \delta_{lL}\delta_{i1}R_{Lm}, \qquad R_{Lm} = \frac{(-i)^L}{2^L}\sqrt{\frac{(2L)!}{(L+m)!(L-m)!}}. \tag{7}$$

Since translational coefficients in the new coordinate system become diagonal in m, expansion coefficients in (5) and (4) with different values of the azimuthal number become independent and can be solved separately. In what follows we will also neglect the cross-polarization terms, described by coefficients $B^{l'm}_{lm}$, which are usually much smaller than $A^{l'm}_{lm}$ [9]. Then, we need only to be concerned with the coefficients b_{lm} and (5).

3 Bi-spheres

In order to understand qualitatively the effect of the coupling on the field distribution, we will first consider a case of a bi-sphere and solve (5) in the so-called single-mode approximation neglecting interaction between modes with different l numbers (see details in [9, 19]). Scattering coefficients for both spheres in this approximation become

$$b_{0_{l,m}}^{(1,2)} = \frac{1}{2}\delta_{lL} R_{lm}\left[\frac{1}{\alpha_L^{-1} - A_{Lm}^{Lm}(\mathbf{r}_2 - \mathbf{r}_1)} \pm \frac{1}{\alpha_L^{-1} + A_{Lm}^{Lm}(\mathbf{r}_2 - \mathbf{r}_1)}\right], \tag{8}$$

where the lower index 0 indicates that this approximation corresponds to the zero order in the inter-mode coupling coefficients. Substituting these expressions into (3), one obtains an expression for the scattered field:

$$\mathbf{E_s} = \frac{1}{2}\sum_m R_{Lm}\left[\frac{\mathbf{M}_{Lm}(\mathbf{r}-\mathbf{r}_1) + \mathbf{M}_{Lm}(\mathbf{r}-\mathbf{r}_2)}{\alpha_L^{-1} - A_{Lm}^{Lm}(\mathbf{r}_2 - \mathbf{r}_1)} + \frac{\mathbf{M}_{Lm}(\mathbf{r}-\mathbf{r}_1) - \mathbf{M}_{Lm}(\mathbf{r}-\mathbf{r}_2)}{\alpha_L^{-1} + A_{Lm}^{Lm}(\mathbf{r}_2 - \mathbf{r}_1)}\right], \tag{9}$$

which gives a clear physical picture of the phenomenon under consideration. This expression describes an optical response with resonances at two sets of frequencies: one is given by the zeroes of $\alpha_L^{-1} - A_{Lm}^{Lm}$ and the other by the zeroes of $\alpha_L^{-1} + A_{Lm}^{Lm}$. The role of coupling between spheres is described by translation coefficients A_{Lm}^{Lm}, which shift the resonant frequency from the single-sphere values by *different amounts for different values of* m. As a result terms with different azimuthal numbers resonate at different frequencies so that one ideally could expect $2(L+1)$ resonance peaks in the optical response of the system, contrary to the coupled-mode theory expectation of just two resonances. The actual number of observed peaks depends on the relation of spectral intervals between adjacent peaks to their widths.

One can notice that the numerators of terms in (9) corresponding to different values of m have a form of symmetric and anti-symmetric combinations of respective single-sphere modes, which is typical for the coupled-mode approaches. This result follows, of course, from the diagonal in m nature of the translation coefficients, which play the role of the perturbation matrix in this approach. In other words, WGMs, defined in the coordinate system XYZ, form a correct basis, in which the perturbation is diagonal in the space of the degenerate states.

More accurate treatment of the situation requires taking into account terms which are non-diagonal in the angular number l. These terms describe coupling between non-degenerate WGMs in different spheres, which result in shifting frequencies of the resonances as well as in changes of their widths [9, 19]. One can describe effects of these terms within a perturbation approach, where an effective small parameter is the expression $\alpha_l A_{lm}^{Lm}(\mathbf{r}_2 - \mathbf{r}_1)$. The nature of the smallness of this parameter is not trivial: the non-diagonal in l elements of the translation coefficients are not by themselves small; actually, they grow with l. The product $\alpha_l A_{lm}^{Lm}$ is nevertheless small because α_l decreases as its pole moves away with increasing l from the frequency of the main mode with $l = L$.

In the case of a bi-sphere the first-order correction to the scattering coefficients $b_{1l,m}^{(1,2)}$ with $l \neq L$ can be presented in the following form:

$$b_{1l,m}^{(1,2)} = \frac{1}{2} b_{0Lm}^{(1)} \alpha_l A_{lm}^{Lm}(\mathbf{r}_2 - \mathbf{r}_1)\left[\frac{1}{1-\alpha_l A_{lm}^{lm}} \mp \frac{1}{1+\alpha_l A_{lm}^{lm}}\right], \tag{10}$$

where $b_{0Lm}^{(1)}$ is given in (8) and we neglected second-order corrections to the denominator, which would modify real and imaginary parts of the resonance frequency of the lth mode obtained in the single-mode approximation. This perturbation theory would break down if in the vicinity of the main resonant frequency, x_L, modes with several other values of l would also have their resonances with high enough values of Q. This situation is quite possible at least for some values of L, as was pointed out in [9, 19]. If this happens, all resonant modes must be treated exactly, while the rest of them can still be treated perturbatively (see details in [9, 19]).

The bi-sphere problem can also be solved numerically exactly using, for instance, matrix inversion. The number of terms with different angular numbers which have to be taken into account is determined from convergency of the procedure. For computations we chose $L = 29$ and a spectral interval in the vicinity of the resonance frequency of the mode with radial number $s = 1$, $x_{29,1} = 21.46$. We checked (for a few representative frequencies) that the convergency is achieved if one takes into account all $l \leq 40$, and that the terms with $l > L$ do not change the results significantly. Therefore, in order to shorten computational time we carried out most of the calculations taking into account all $1 < l \leq L$. Including all coefficients with $l < L$, is necessary to insure that we include all possible resonance modes that can significantly affect the results.

Using the found scattering coefficients we calculate expansion coefficients of the internal field $d_{l,m}^{(i)}$:

$$d_{lm}^{(i)} = \frac{i}{x}\frac{1}{j_l(x)[nxj_l(nx)]' - j_l(nx)[xj_l(x)]'} b_{lm}^{(i)}, \tag{11}$$

where $j_l(x)$ is the spherical Bessel function and $[zj_l(z)]'$ means differentiation with respect to z. Knowing the coefficients $d_{lm}^{(i)}$, we can find the total energy of the field concentrated inside spheres as a function of frequency, which is best suited to characterize the optical response of our system in the spectral range of high-Q WGMs [9]. Figures 2a

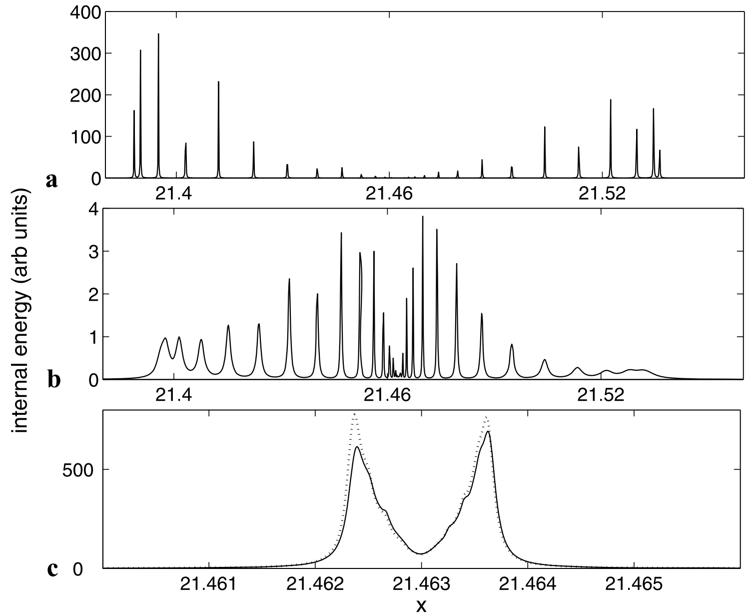


Fig. 2 The internal energy of a bi-sphere as a function of dimensionless frequency, x. **a** Single-mode approximation, $d = 0$; **b** numerical calculations including all modes with $l \leq 29$, $d = 0$; **c** the double-peak spectrum obtained for $d = 0.28R$: *solid line*—numerical multi-mode calculations, *broken line*—single-mode approximation

and b present the spectra of the internal energy obtained by two procedures: exact multi-mode numerical computation and calculations based on the single-mode approximation (9). One can see that while the single-mode model reproduces the multi-resonance optical response, it deviates strongly from exact numerical calculations in number, positions, and heights of the respective peaks. Comparing the latter for Figs. 2a and b, we can conclude that the inter-mode coupling significantly reduces Q-factors of the respective resonances.

With increasing distance between spheres the distance between resonance peaks decreases because of the reduced coupling, and adjacent resonances start overlapping. At a certain value of the inter-sphere gap d, the two-peak structure seen in Fig. 2c emerges. These peaks, however, cannot be identified with frequencies of bonding and anti-bonding states of the coupled-mode theory even though they are well described by the single-mode approximation. Indeed, the double-peaked spectrum in our calculations arises as a result of overlapping of multiple resonances when decreased coupling pushes them all toward the single-sphere resonance, making spectral separations between them smaller than their radiative widths. In the absence of radiative broadening all these resonances would have maintained their individuality for an arbitrarily weak coupling. Respectively, the positions of the emerging peaks are determined by an interplay of the m dependence of the radiative lifetimes of individual resonances, the coupling parameters A_{Lm}^{Lm}, and the excitation parameters R_{Lm} and cannot be directly related to the overlap integral of the coupled-mode theory. Moreover, the widths of the peaks in our calculations are due to inhomogeneous broadening caused by the overlap of unresolved resonances with different m rather than due to homogeneous radiative broadening of individual resonances, contrary to the standard assumption of the coupled-mode approaches.

In order to further elucidate the role of the inter-mode coupling we present the absolute values of the coefficients of the internal field $d_{lm}^{(i)}$ for all values of $0 < l \leq 29$ and respective values of $|m| \leq l$. In order to visualize the results we present pairs l, m as a one-dimensional array ordered according to the following rule: $(29, -29), (29, -28), \ldots, (29, 0), (29, 1), \ldots, (28, -28), \ldots$ and plot coefficients d_{lm} versus a number of the respective pair in this array. Figure 3 presents the results of these calculations for $l \leq 25$ for one of the resonance frequencies, $x = 21.415$. One can see that the largest values of the coefficients correspond to the main angular number $l = 29$, which as a function of m shows two large symmetric peaks for $m = \pm 4$. The positions of the peaks are determined by the dependence of the resonant frequency on the azimuthal number: the resonance at the chosen frequency results from the component of the internal field with $|m| = 4$. Besides main coefficients corresponding to $l = 29$ there are present other coefficients as well, which, however, have much smaller values. The second largest coefficient corresponds to $l = 25$, which is in agreement with the fact that a frequency of the mode with this value of l and with the radial number $s = 2$ is almost in resonance with our main mode, $l = 29$, $s = 1$ [9]. The effect of this resonance is still small because of the low Q value of the $l = 25$, $s = 2$ mode, and not sufficient spectral overlap between the two. Nevertheless, the inter-mode coupling results in significant deviations from predictions of the single-mode model, while one can expect that in this case an accurate description of the electromagnetic field in our system can be obtained with the first-order expression (10). The situation can be different, however, when one considers

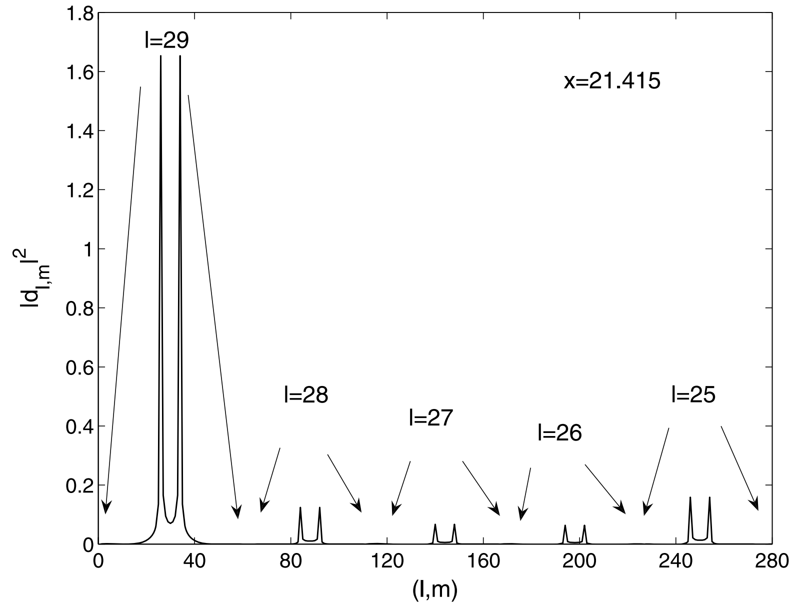
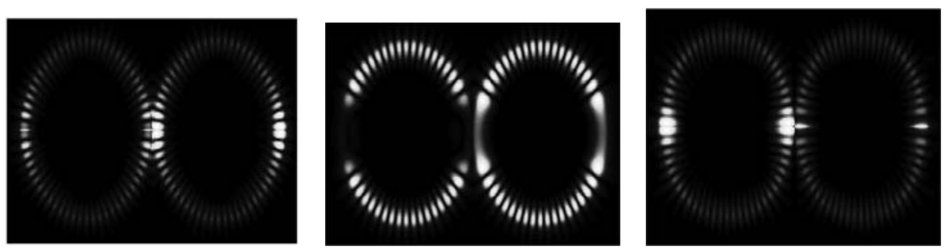


Fig. 3 Internal field coefficients for all values of l and m for $l \leq 25$. *Arrows* indicate the regions corresponding to coefficients with given l and varying m



a **b** **c**

Fig. 4 Distributions of the field intensity at several resonant frequencies. **a** For $x \approx 21.40$. **b** For $x \approx 21.46$. **c** For $x \approx 21.50$

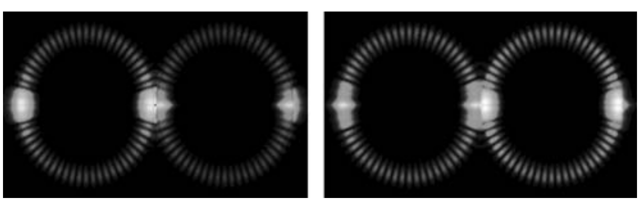


Fig. 5 Distribution of the field in the regime of weak coupling for the lower frequency (*left-hand figure*) and the higher frequency (*right-hand figure*) resonances of the double-peak spectrum shown in Fig. 2c

coupling between other fundamental modes. For instance, the TE mode with $l = 39$, $s = 1$ has a very strong spectral overlap with another TE mode with relatively high Q-factor ($l = 34$, $s = 2$) [9, 19], which results in a strong resonant interaction between the two modes. Therefore, the perturbative approach of (10) breaks down in this case and one can expect strong deviations from the single-mode approximation, not to mention the coupled-mode approach.

To conclude our discussion of the bi-sphere we present calculations of the distribution of the intensity of the field on the surfaces of the spheres in the XZ plane. These distributions were calculated at three different resonance frequencies from those seen in Fig. 2 and are shown in Figs. 4a, b, and c. One can see that the field distribution in the spheres drastically changes from one frequency to another, and is very far from resembling bonding and anti-bonding orbitals of the mode-coupling theory. In order to reinforce the point that even in the weak-coupling regime the two resonances do not correspond to the bonding and anti-bonding orbitals, we also constructed the distribution of the field for frequencies corresponding to both peaks shown in Fig. 2c. The results shown in Fig. 5 demonstrate that even in this case the resulting distribution of the field cannot be described as linear combinations of two fundamental modes.

4 Multi-sphere chains

In order to treat the case of N spheres exactly one needs to solve numerically the system of $2 \times l_{\max} \times N$ equations (4) and (5). Neglecting cross-polarization terms the number of equations is reduced by half, but still for a large number of the spheres it is too expensive. We have carried out these calculations for $N = 3, 4, 5$ keeping the number of modes with different angular numbers up to $l_{\max} = L$. In Figs. 6, 7, and

Fig. 6 $N = 3$

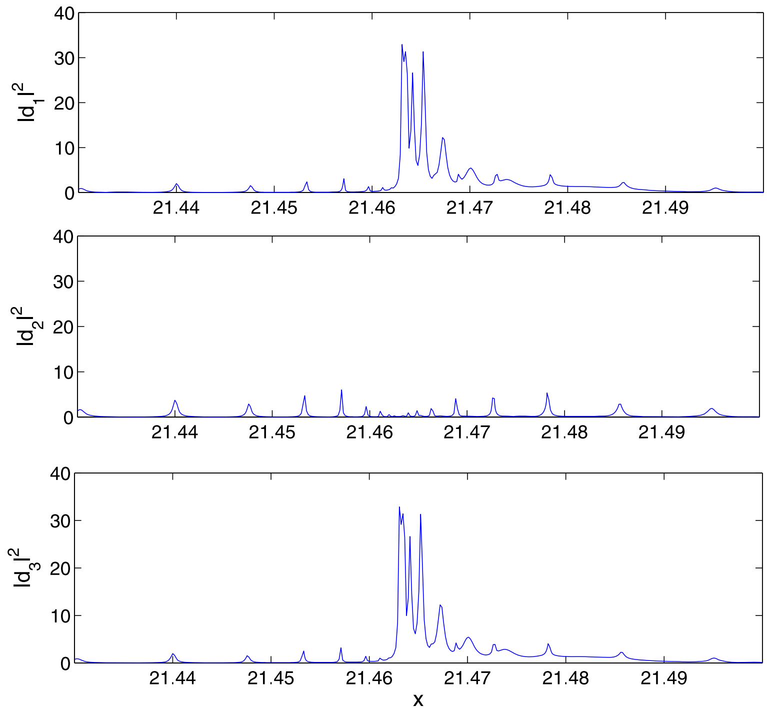
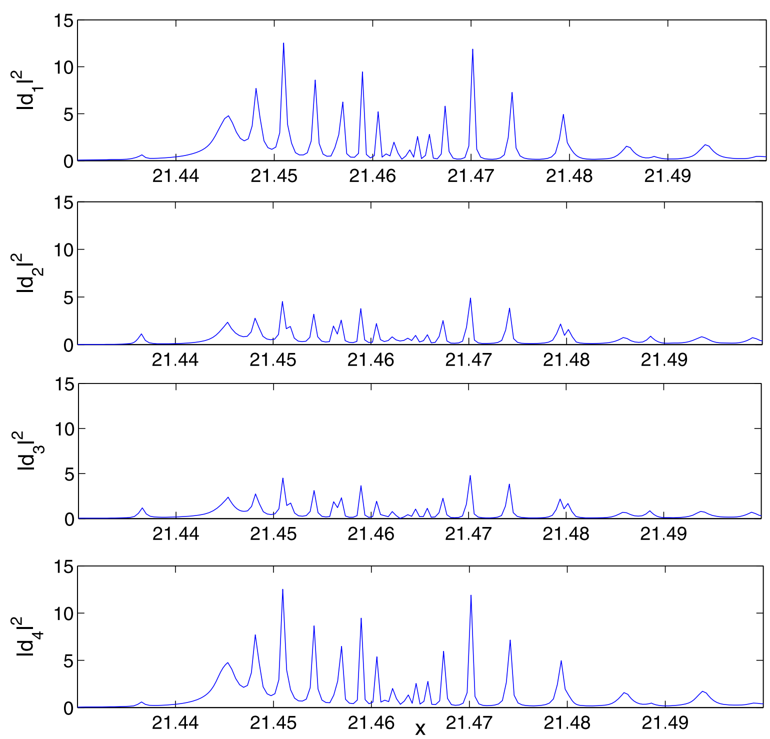


Fig. 7 $N = 4$



8 we present $|d_i^2| = \sum_{l,m} |d_{lm}^{(i)}|^2$, which is the sum over all l and m of the absolute values of the expansion coefficients of the internal field for all spheres in each of these chains.

These figures show not only positions of the resonance frequencies, which are obviously the same for all spheres in a chain, but also the distribution of the internal field between the spheres. There are several interesting features seen in these figures that deserve a discussion. First of all, one could expect that the number of peaks must increase with the number of spheres in the chain as each new structural element results in additional splitting of the frequencies; we, however, do not see it in the figures. Explanation for the fewer than expected number of resonance maxima is quite simple—new resonances must occupy the same frequency range regard-

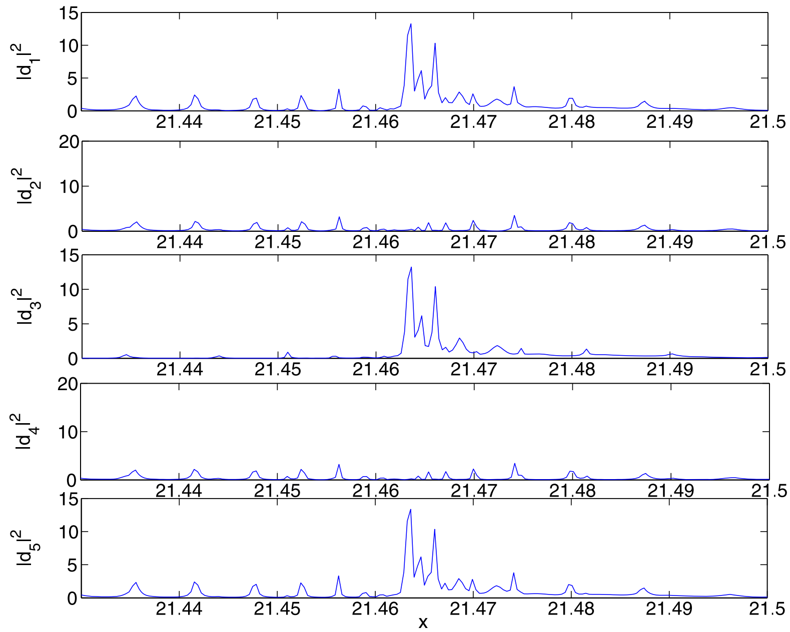


Fig. 8 $N = 5$

less of the number of the spheres in the chain. Therefore, new additional spheres result in an increased density of the resonant frequencies and, since all these resonances have finite widths, many of them overlap forming single inhomogeneously broadened peaks. What is more interesting is that the number of resonances as well as their distribution over the frequency interval differs significantly between chains with even and odd numbers of spheres. Indeed, for $N = 3$ and $N = 5$ the strongest resonances are found in the vicinity of the single-mode frequency $x_{29,1}$ with one strongest maximum being exactly at this frequency. On the other hand, in the cases of $N = 2$ and $N = 4$, the resonances are spread over a larger frequency interval with the vicinity of $x_{29,1}$ being essentially devoid of the peaks. Another difference between odd and even chains is the distribution of the field between the spheres. For instance, the peak corresponding to the single-sphere resonance shows the same largest height in all odd-numbered spheres in the cases of $N = 3, 5$, while it does not appear at all in the even-numbered spheres. A qualitatively similar pattern can be noticed for two other main peaks to the right of $x_{29,1}$. At other frequencies, the intensity of the field demonstrates a different behavior; for instance, the largest peak between $x = 21.45$ and $x = 21.46$ decreases gradually from the first sphere to the third, and then goes up again reaching the original value at the fifth sphere. In the case of $N = 4$ the situation is qualitatively different: now spectra are almost identical for first and last spheres and for spheres numbers 2 and 3 with the first of these pairs showing a much stronger field intensity.

In order to understand the obtained results, one can rely on a nearest-neighbor coupling approximation, which was employed in [19] to study normal modes of chains of microspheres. This approximation works well in the case of high-Q whispering gallery modes because the translation coefficients $A_{lm}^{l'm'}(\mathbf{r}_i - \mathbf{r}_j)$ for $l, l' \gg 1$ fall off with the distance between the spheres very fast. In this approximation equations for the scattering coefficients can be written down as

$$\frac{1}{\alpha_l} b_{lm}^{(i)} - \sum_{l'} A_{lm}^{l'm} \left[(-1)^{(l+l')} b_{l'm}^{(i-1)} + b_{l'm}^{(i+1)} \right] = \eta_{lm}^{(i)}, \qquad (12)$$

where $A_{lm}^{l'm}$ is a translation coefficient between the ith and $(i+1)$th spheres. The factor $(-1)^{(l+l')}$ in front of $b_{l'm}^{(i-1)}$ reflects the symmetry of the translation coefficient upon inversion: $A_{lm}^{l'm}(\mathbf{r}_1 - \mathbf{r}_2) = (-1)^{(l+l')} A_{lm}^{l'm}(\mathbf{r}_2 - \mathbf{r}_1)$ (this factor was omitted in [19], which resulted in a wrong dispersion relation in the case of coupling between modes with angular numbers of different parity). This equation should be complemented by two boundary conditions:

$$b_{lm}^{(0)} = 0, \qquad b_{lm}^{(N+1)} = 0, \qquad (13)$$

where the first expression takes into account that there are no spheres to the left of the first one, and the second reflects the absence of a sphere to the right of the last one.

Solutions to (12) can be presented as a linear combination of respective normal modes, $B_{lm}(i)$, satisfying the boundary condition (13). To qualitatively understand the results of simulations shown in Figs. 6, 7, and 8, it is sufficient to solve (12) in the single-mode approximation, in which case the normal modes are given by

$$B_{Lm}^{k}(i) = \tilde{B}_{Lm}^{k} \sin\left(\frac{\pi i k}{N+1} \right), \qquad (14)$$

where $k = 1, 2, \ldots, N$ enumerates various normal modes, and the amplitudes $\tilde{B}_{Lm}^{k}$ are determined by the incident field coefficients (7) and are equal (with accuracy to a normalization factor) to

$$\tilde{B}^k_{Lm} = \sin\left(\frac{\pi k}{N+1}\right)\frac{R_{Lm}}{\alpha_L^{-1} - A^{Lm}_{Lm}\cos(\frac{\pi k}{N+1})}. \quad (15)$$

Poles of these amplitudes given by the equation

$$\alpha_L^{-1} - A^{Lm}_{Lm}\cos\left(\frac{\pi k}{N+1}\right) = 0 \quad (16)$$

determine N complex-valued eigenfrequencies of the chain. However, as was pointed out already, not all of these eigenfrequencies show up as resonance peaks in the spectrum, because many peaks with differing values of m are too close to each other so that they overlap and form single inhomogeneously broadened peaks. Despite this, (14), (15), and (16) are very useful in understanding the results shown in Figs. 6, 7, and 8.

First of all, let us note that in the case of odd N, there always exists a mode characterized by $k = (N+1)/2$, for which $\cos(\pi k/(N+1)) = 0$. The eigenfrequency of this mode coincides with the resonance frequency of the WGM in a single sphere, and *is independent of the azimuthal number m*. This means that for this, and only for this, mode the distribution of the field in each sphere of the chain reproduces the field of the single-sphere fundamental mode. *Thus, if one wishes to arrange propagation of an undistorted fundamental mode through a chain of spheres, one needs to deal with a structure consisting of an odd number of elements and work at the frequency of the single-sphere resonance.* The spectral width of the pulses that can propagate along such a chain with little distortion is determined by a spectral interval between the main single-sphere peak in Fig. 8 and the closest adjacent peak. The spatial profile of this mode is given by the expression $B^{(N+1)/2}_{Lm}(i) \propto \sin(\pi i/2)$, which is maximum at odd-numbered spheres, and turns to zero at even-numbered ones, explaining the behavior observed numerically, Figs. 6 and 8. Modes with smaller or larger values of k are characterized by a smoother dependence of the sphere number, as is seen in the same figures. In the case of even N the mode with $k = (N+1)/2$ does not exist, and this why there are no peaks in the vicinity of the single-sphere resonance in Fig. 7.

Another feature of the case $N = 4$, namely the similarity in the field distributions between spheres with $i = 1$ and $i = 4$, as well as between spheres with $i = 2$ and $i = 3$, can also be understood from (14). One can see that the distribution of the field seen in Fig. 7 is associated with modes characterized by $k = 2$ and $k = 3$. The amplitudes of these modes at their respective resonances are proportional to $\sin(2\pi/5)$, which is larger than the amplitudes of modes with $k = 1$ and $k = 4$ proportional to $\sin(\pi/5)$. Therefore, the observed spatial distribution of the field, which is a sum of contributions from all four modes, more closely resembles the pattern characteristic for $k = 2, 3$. Thus, we see that the single-mode approximation can account for the spatial distribution of the field in linear chains of spheres, but the number, exact positions, and widths of the resonance peaks in their optical response can only be predicted on the basis of the full multi-mode theory.

5 Conclusion

In this paper we carried out a careful study of propagation of fundamental modes in a linear chain of spherical microresonators. We pointed out that the coupled-mode type of arguments or simple perturbation theory calculations do not apply to this situation because of the degeneracy of single-sphere resonances. We also showed that the multi-sphere Mie formulation provides not only a tool for accurate numerical simulations of the properties of the chain, but can also be used as a natural foundation for developing a perturbation theory, in which the degeneracy of uncoupled modes is taken into account automatically. We carried out accurate numerical calculations of the chains consisting of two, three, four, and five spheres, and complemented them by an approximate analytical analysis based on single-mode and nearest-neighbor approximations.

In the case of a bi-sphere our analysis revealed that the electromagnetic energy stored in the spheres is characterized by a much richer spectrum with multiple resonances than just two peaks predicted by coupled mode type approaches. These multiple resonances correspond to modes with different values of the azimuthal number, which are excited in the system due to the violation of the complete spherical symmetry. The exact number of the resonance peaks and their positions depend on the coupling strength and are determined by an interplay between the inter-resonance spacing and the widths of the resonances. Even in the case of a weak coupling, when only two peaks in the spectrum survive, we argue that these peaks do not correspond to bonding and anti-bonding modes of the coupled-mode theories.

Considering chains with $N > 2$ number of spheres we found that there exists a significant difference between the behavior of chains with N even and N odd. We showed that qualitatively all these differences can be accounted for within the framework of the simple nearest-neighbor single-mode approximation. In the case of chains with odd N we found that there exists a mode with a frequency which does not depend on the azimuthal number and coincides with the frequency of the single-sphere resonance. This mode can be used for propagating the single-sphere fundamental resonance along the chain with smallest possible distortion caused by inter-sphere coupling.

The question arises, however, of how our results agree with observations of bonding and anti-bonding orbitals with two split frequencies reported in many experimental works on bi-spheres. One needs to separate these experiments into two groups. In the experiments of [7] or [20] the observed modes were true *normal modes* of the bi-spheres, characterized by a well-defined azimuthal number m. These modes are not *strongly coupled fundamental modes* in the sense described above, so they are not the subject of this paper. The same is true for multi-sphere experiments presented in [21, 22]. The second type of experiment, such as described in [13, 23], deals with modes of the spatial configuration similar to those discussed here. It should be noted, first of all, that these experiments dealt with spheres of different diameters meaning that the resonant single-sphere modes corresponded to different azimuthal numbers l. Our calculations did not cover this situation, which is more complicated. Nevertheless, when interpreting this type of experiment one should be aware that an observed two-peak structure can appear as a result of the collapse of the multi-peak response demonstrated in this paper rather than as a splitting of a single-sphere mode into binding and anti-binding orbitals of the coupled-mode theory. Our calculations show that the two-peak spectrum might represent an inhomogeneously broadened envelope of unresolved multiple resonances, each with its own Q-factor. As a result, it would be a mistake to relate the spectral width of these peaks to radiative lifetimes and their spectral separation to the strength of optical coupling.

We are not aware of experimental works in which the observation of coupling of fundamental modes would have been attempted in a multi-sphere chain. We believe it would be of great interest to observe a propagation of the fundamental mode along an odd-numbered chain of spheres via the 'non-distorting' collective mode, discussed in the paper.

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