Low order modes in microcavities based on silicon colloids

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Abstract: Silicon colloids based microcavities, with sphere size between 1 and 3 micrometers, have been synthesized and optically characterized. Due to both the small cavity volume and the high refractive index of silicon we are able to tune resonances with extremely low mode index, whose electric field distribution resembles those of electronic orbitals. The value of some parameters such as quality factor Q, effective mode volume, and evanescent field have been calculated for several modes. This calculation indicates silicon colloids can be a serious strategy for developing optical microcavities where may coexist both optical modes with large evanescent fields useful for sensing applications, as well as modes with high Q/V ratio values, of the order of $10^9(\lambda/n)^{-3}$.

OCIS codes: (140.3945) Microcavities; (040.6040) Silicon.

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1. Introduction

Silicon is a key material in electronics. All semiconductor industries are based on the planar technology with a top-down development strategy, i.e.: from silicon wafers tens of cms large, millions of identical devices are developed simultaneously. The integration capability has grown so rapidly that it is reaching the so called Sound Barrier for Silicon Technology [1]. Silicon is also a key material in photonics. A large amount of photonic systems and devices based on silicon such as 2D [2] and 3D [3] photonic crystals, multiplexers [4], to name a few, have been developed for the last few years. A considerable number of these devices are Whispering Gallery Modes (WGM) based optical microcavities [5,6] with ultra high quality factor (Q) values (Q~10^8) made of silicon and also of other materials like InGaAsP, GaAs, SiO_2, etc [7–9]. These systems, although with extremely high performance values, are still large in size (several tens of micrometers) because they all use low refractive index materials (SiO_2) as the cavity medium. Recently some of us reported on a completely new type of silicon structures. Chemical Vapor Deposition techniques (CVD) allow the formation of polydisperse silicon micro and nanoparticles, with size ranging from 0.5 to 5 micrometers, and with a perfect spherical shape [10]. We call them silicon colloids or silicon microspheres. Because of their surface perfection, their spherical topology and their high refractive index value, silicon colloids work very well as optical microcavities in the IR region. Here we report on the optical properties of microcavities based on silicon colloids with different particle size values. The small size of the cavities allows the detection of Mie resonances with very low modal numbers, whose electromagnetic (EM) field distribution resembles the electronic density distribution in atomic orbitals. We report on the processing and optical characterization of silicon colloids based microcavities of several sizes, from 1 to 2.5 µm. It is important to stress that the light scattering spectra of microcavities can be fitted very well in all the measured range by using Mie theory, and taking the cavity diameter as the only fitting parameter. In all cases, due to both the small size of colloids and the high refractive index of silicon, very low modal number modes can be detected in the near-IR region.

2. Silicon colloids synthesis and measurement procedure of their optical properties

Silicon colloids are obtained by chemical vapor deposition (CVD) techniques using disilane as a precursor gas [10]. Under controlled chemical reaction conditions, silicon colloids nucleate and grow in disilane gas [11] similarly to the growing process of silica colloids in a liquid solution containing appropriate precursors [12]. Thanks to surface tension forces the colloidal particles become highly spherical with a very smooth surface. This method allows synthesizing amorphous and polycrystalline silicon microspheres, depending on the decomposition temperature of the precursor. For this work, polycrystalline microspheres were chosen. The fabricated microspheres are polydisperse with diameter values from 0.5 to 5 micrometers [10].

The optical characterization of the silicon spheres has been performed by measuring their transmittance spectrum. For this purpose, isolated spheres with different sizes have been placed on a glass substrate. The glass substrate barely affects the characterization of the silicon spheres due to the high refractive index contrast between glass and silicon (the refractive index for glass is 1.5 and for silicon is around 3.5 for the wavelength range under study). Micromanipulation techniques have been developed and needle shaped tools of different materials have been fabricated for pick and place operation and for the accurate positioning of the spheres on the substrate. The spheres were selected based on their size, by using an optical microscope at 1000x magnification. This method allows us to know an approximated size of the sphere, whereas a nanometric resolution size value can be determined from the transmittance spectrum fitting to Mie theory, as we will see below.

The transmittance spectra of the silicon spheres were measured by a Fourier Transform Infrared Spectrometer Bruker IF 66/S with a coupled microscope for the wavelength range from 1 to 4 μm, and a Fourier Transform spectrometer Bruker Equinox 55 with a coupled microscope for the wavelength range from 3 to 16 μm.

3. Resonating modes in silicon colloid microcavities

Spheres with different diameter, *d* from 1 to 3 μm have been characterized. The transmittance spectra measured for three of them are shown in Fig. 1 (a), (b) and (c) (black curves). Each dip in transmittance corresponds to a MIE mode. The labels under each mode indicate the type of mode and its order. Two types of resonating modes are identified: *a* modes or transverse magnetic (TM), where the magnetic field is parallel to the surface of the sphere, and *b* modes or transverse electric (TE), where the electric field is parallel to the surface of the sphere. The order of the mode is indicated by the two sub-indices that correspond to the number of maximums of the electric field intensity in the half sphere perimeter and in the radial direction, respectively [13]. The measured spectra are compared to the simulated optical response of silicon microspheres, calculated by Mie theory [14,15] (red curves). The refractive index value of silicon, as function of the wavelength, is a well known parameter [16], and the sphere size is the only fitting parameter. This allows to obtain high precision diameter values (better than 0.5%), that had been previously estimated by optical microscopy means. These values are 1.05 μm, 1.85 μm and 2.49 μm for the microspheres of Fig. 1 (a), (b) and (c) respectively. The good agreement between experiment and simulation in all the measurement range demonstrates: a) the highly spherical perfection and the smoothness of the cavity, and b) the cavity is made of polycrystalline silicon.

One of the most outstanding aspects of silicon colloid microcavities is the fact that they can sustain low order resonating modes. For example, in Fig. 1(a) (sphere with d = 1.05 μm), the fundamental modes $b_{1,1}$ and $a_{1,1}$, appear at wavelength values around 4 μm and 3 μm

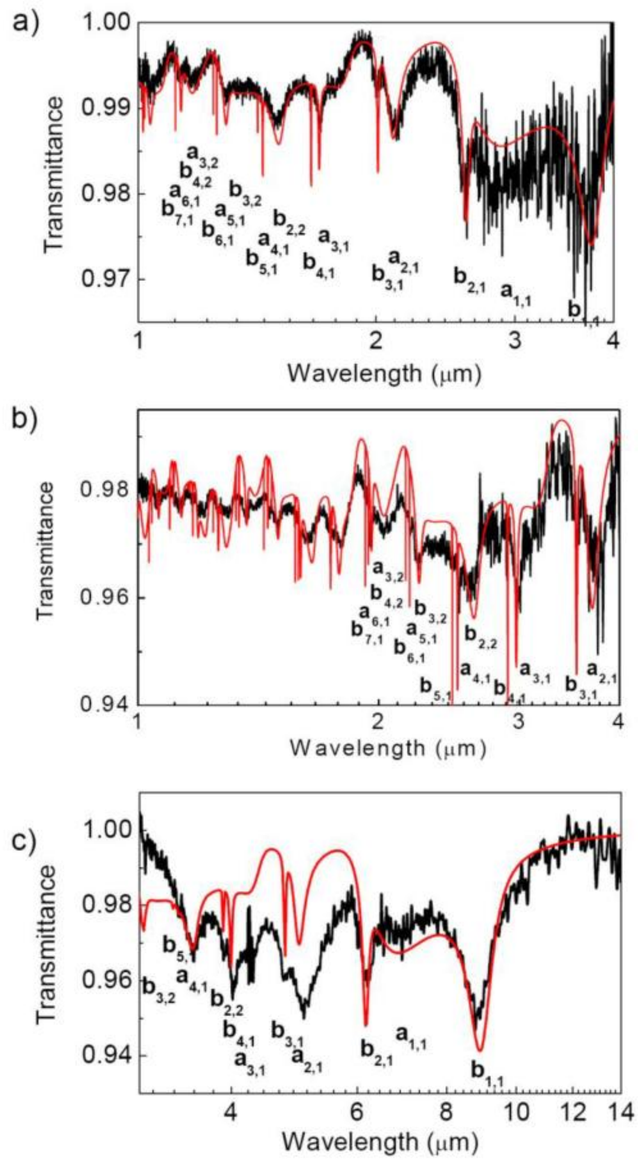

Fig. 1. Measured optical transmittance spectra (black curves) of silicon spheres with diameter a) 1.05 μm, b) 1.85 μm and c) 2.49 μm. The simulated optical response of the spheres are also plotted (red curves). The dips in transmittance correspond to Mie modes. Some of them are labeled under their corresponding dip indicating the type and order of the mode.

respectively, although they are still quite noisy because they are at the limit of the measurement range of the spectrometer. Figure 1(c) shows more clearly the fundamental modes for the 2.49 μm diameter microsphere. In this case, because of the bigger size of the cavity, $b_{1,1}$ and $a_{1,1}$ are located at larger wavelengths, at about 9 μm and 7 μm respectively.

Although the dip position of the resonances is well fitted by theory, some transmittance amplitude discrepancies can be detected. They may be ascribed to the influence of the substrate used in this experiment. Here, at variance to the former samples, we use as substrate a pellet made of pressed potassium bromide (KBr) powder that is transparent to the infrared radiation above 4 μm. However, KBr is a highly hygroscopic material and the adsorbed water shows absorption features in the spectral region between 3 μm and 5 μm where the discrepancies are more visible. Also unwanted scattering effects from the KBr powder may contribute to the differences between experiment and theory.

The key factor for silicon colloids to be able to sustain low order modes is the high refractive index of silicon (around 3.5 for the near IR wavelength range). Typical dielectric spheres made of silica or polystyrene can only sustain higher order modes because of their low refractive-index contrast relative to air or water environments [17]. In order to better understand the importance of this fact, Fig. 2 shows the simulated scattering cross section of two 2 μm diameter microspheres, the first one (red curve) is made of silica and the second one (black curve) is made of silicon. Both spectra are represented as a function of the wavelength λ (top axis) and of the size parameter, expressed as $\alpha = \pi d/\lambda$ (bottom axis). We found, at variance to silicon, that silica spheres do not present well defined resonant modes at low size parameters, although we can guess their existence from the small ripples of the spectra. In contrast, silicon colloids based microcavities show well defined resonances even in the case of the lowest order modes, as indicated in the figure.

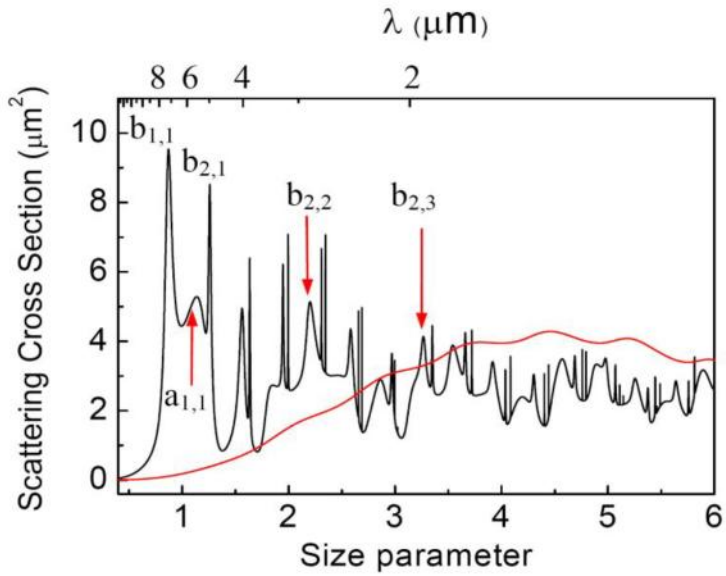

Fig. 2. Simulated scattering cross section of 2 μm diameter silicon (black curve) and silica (red curve) spheres. The peaks correspond to Mie modes, or WGM's. While the silicon sphere shows resonances for low size parameters (see for instance the low order modes $b_{1,1}$ and $a_{1,1}$, etc. indicated in the figure) the silica sphere yields a broad rippled structure.

According to Mie theory the resonances of microspheres are located at specific values of the size parameter. This is very important because we can tune a certain mode at the wavelength of interest by only selecting the appropriate sphere diameter, taking into account some slight variations that come from the refractive index dispersion at infrared wavelengths. For instance, the mode $b_{4,1}$ appears at a wavelength of about 3 μm for the 1.85 μm diameter microsphere [Fig. 1 (b)], but it is located at wavelengths in the telecommunication region, i.e. around 1.5 μm for the 1.05 μm diameter microsphere [Fig. 1 (a)].

We have measured and analyzed the optical transmittance spectra of many silicon microspheres with different diameters. Some of them are shown in Fig. 3(a). They are plotted as a function of the size parameter that was calculated for each microsphere by using the diameter values obtained from fitting the curves to Mie theory, as it has been described above. The fitted curves and the obtained diameters are shown in Fig. 3(b), and the resonating modes are indicated under their corresponding dip. The larger the sphere diameter, the bigger the scattering cross section, and consequently, the more pronounced the transmittance dips.

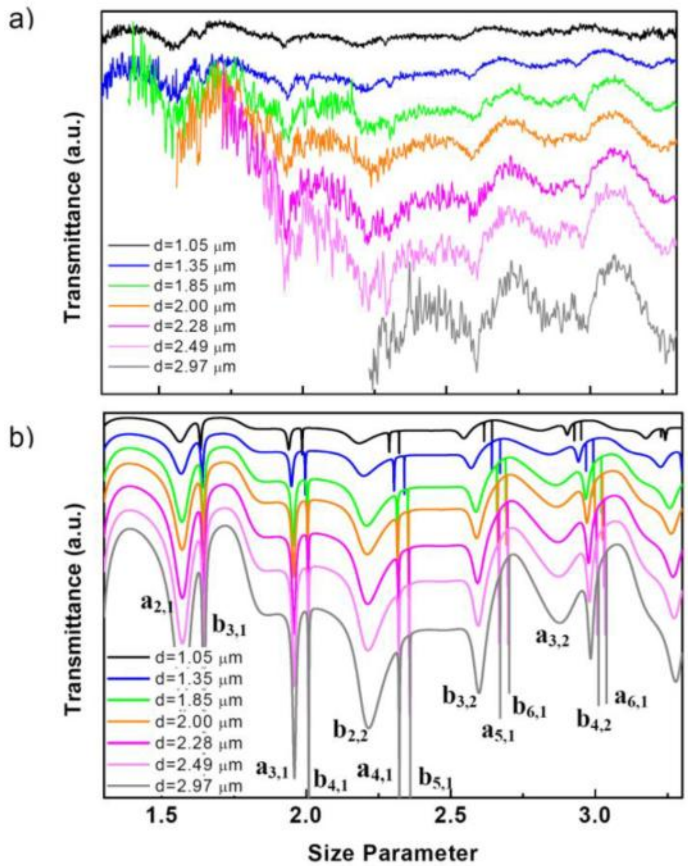

Fig. 3. (a) Measured transmittance spectra of silicon spheres with different size. (b) Fitted curves to the transmittance spectra of (a). The obtained diameters, d, are indicated. The dips in transmittance correspond to Mie modes, whose order is indicated by the labels under the corresponding dip.

Figures 3 (a) and (b) illustrate the fact that the scattering spectra are fairly similar for all size microcavities when plotted in units of the size parameter value. Small deviations, which are attributed to the refractive index dispersion, are more easily discernable for higher size parameter values and for smaller spheres because the modes are located in these cases at shorter wavelengths, where the dispersion of the refractive index is more pronounced. This is the case, for instance, of modes $a_{3,2}$ and $b_{4,2}$ of the smallest spheres.

4. Light confinement in silicon colloid microcavities for low order modes

Silicon colloids may facilitate high-quality-factor optical microcavities [18] with strong light confinement effects [10]. The electromagnetic energy localized in a microsphere at resonant frequency values is given by the quality factor (Q) of the resonance mode. Q is defined as Q=2π(Total stored energy)/(Energy loss per cycle). It can also be expressed as the ratio between the resonant frequency f_c and the full width at half-maximum of the mode. The higher the Q, the more energy is confined in the sphere.

When the EM energy is resonating in a sphere, an important part of this energy is confined inside the sphere and another part can be detected in the surrounding area. Figure 4 shows the

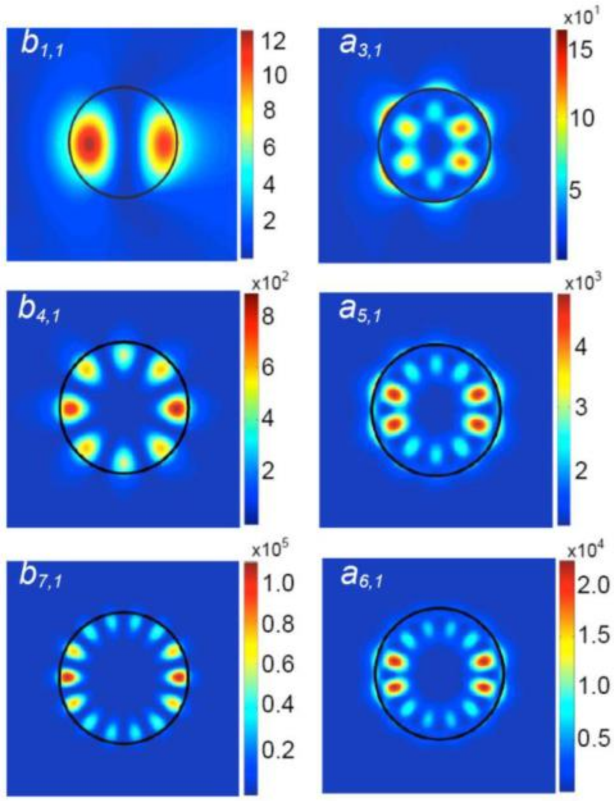

Fig. 4. Electric field intensity distribution inside and nearby a silicon sphere in the resonance plane for different resonating modes. The amplitude of the electric field is given in atomic units.

spatial distribution of the electric field intensity for different order modes. They were calculated by using a semi-analytical method based upon multiple elastic scattering of multipole expansions (MESME) [19]. Because they are low order modes, their associated distributions are relatively simple, bearing a strong resemblance to the atomic orbitals. This is very important for performing fundamental experiments like those of photonic forces [20], where simple EM distributions may be desirable.

Table 1 shows different calculated parameters that are associated to the light confinement phenomenon for several resonating modes, indicated in the first column. The second column indicates the position of the modes (in units of size parameter). We supposed the sphere diameter is 1.5 μm and the dielectric constant equals to 12 in all the calculated wavelength range. This allows deducing the position of the modes in wavelength, which is written in the 3rd column of the table.

One of the most remarkable characteristics of the resonating modes in silicon colloid based microcavities is their low mode volume (V_m), specially for the lowest order modes that have a value below $1(\lambda/n)^3$ (see for example V_m of mode $b_{4,1}$ in column 5). This parameter was calculated by using equation):

$$V_m = \frac{\int_V \varepsilon(r)\left|E(r)\right|^2 d^3r}{\max\left[\varepsilon(r)\left|E(r)\right|^2\right]}, \tag{1}$$

where ε(r) is the dielectric constant and E(r) is the electric field. The calculation of Eq. (1) presents inherent problems for low modal number modes. For practical reasons we have considered a volume of cubic shape with different edge length values to guarantee the convergence of the integral. Convergence has been obtained when the side of the cube is six times the radius of the microsphere. The microsphere is centered inside the cube. The V_m calculated in this volume can be considered equivalent to the calculation using a bigger volume for modes higher than $a_{3,1}$ as the electric field amplitude generated by the mode decreases considerably with the distance to the microsphere.

Table 1. Size parameter (α), wavelength, Q-factor, normalized modal volume, normalized Q/V_m ratio and percentage of the external electric field in the surrounding area of the sphere calculated as 100*/E^2/$_{external}$/(/E^2/$_{external}$+/E^2/$_{internal}$) for different order modes. The notations internal and external indicate the integration of /E^2/ inside and outside the sphere respectively. The sphere diameter is 1.5 μm.

Mode	α	λ (μm)	*Q*	V_m $(\lambda/n)^3$	Q/V_m $(\lambda/n)^{-3}$	Ext Field (%)
$b_{4,1}$	1.9882	2.37	9.1 x10^2	0.7639	1.184 x10^3	45.80
$a_{6,1}$	2.9862	1.58	2.7x10^4	1.2818	2.149 x10^4	33.53
$a_{9,1}$	3.9759	1.18	4.9x10^6	2.0152	2.476 x10^6	22.48
$a_{11,1}$	4.6219	1.02	1.7x10^8	2.4246	6.846x10^7	18.33
$b_{13,1}$	4.9616	0.95	3.8x10^9	2.6692	1.424 x10^9	8.78

V_m does not increase significantly when increasing the order of the mode. However, the *Q* value (column 4 in Table 1) increases enormously, from 9.1x10^2 for $b_{4,1}$ mode to 3.8x10^9 for $b_{13,1}$ mode. This way, high Q/V_m ratios can be obtained, as illustrated in column 6. These values are of the same order of magnitude, and in some cases even higher than those achieved previously in other types of cavities [21–24]. Therefore, silicon colloids are good candidates for realizing experiments in the framework of quantum electrodynamics. However, such high *Q* values have not been measured so far. This constitutes currently an object of research. It is remarkable that the *Q* factor of the silicon microspheres studied here is of radiative origin and not originated by absorption losses in silicon as the extinction coefficient for silicon is negligible for the wavelength range under study [16].

Although achieving modes with high *Q* value is very interesting, sometimes it is desirable to have lower *Q* modes because in this case the evanescent field, and therefore, the percentage of electromagnetic energy of the mode outside the cavity is larger. Therefore, it favors the photonic interaction between the cavity and any surrounding element, and it can be convenient for sensing applications, etc. In any case, taking into account the scalability of the resonating wavelengths with the sphere size, previously explained, we may have, for instance, either a high evanescent field mode ($b_{4,1}$) centered at λ by selecting a sphere with diameter d_1 or a highly confined mode ($b_{13,1}$) at the same λ by selecting a sphere with d_2. The important aspect is that we have an adjustable *Q* factor ranging seven orders of magnitude by only increasing 2.5 times the diameter of the sphere ($d_2/d_1=\alpha_2/\alpha_1$) thus choosing sphere sizes values of the same order of magnitude.

5. Conclusions

We have measured the optical transmittance spectra in the near infrared of different diameter silicon colloid microcavities, from 1 to 3 micrometers approximately. All of them could be fitted to Mie theory and they show well defined resonating modes, whose position in the spectra scales proportionally to the sphere diameter. Because of the high refractive index of silicon, the colloids can sustain low order modes, such as $b_{1,1}$ and $a_{1,1}$, etc, where the electromagnetic field intensity distribution is very simple and it resembles that of the electronic density in atomic orbitals. Therefore, silicon colloids can be very useful as a platform for studying photonic interactions.

We have also calculated the value of some parameters for several optical modes in silicon colloid microcavities in the framework of quantum electrodynamics. This calculation indicates that silicon colloids can be of application for developing optical microcavities where may coexist both optical modes with large evanescent fields useful for sensing applications, as well as modes with high Q/V_m ratio values.

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