Whispering Gallery Mode for Second-Harmonic Generation in Microresonators

G. Kozyreff (1,2), J.L. Dominguez Juarez (1), **and Jordi Martorell (1,3)**

1) ICFO-Institut de Ciencies Fotoniques, 08860 Castelldefels (Barcelona), Spain

2) ONT, Université Libre de Bruxelles, Campus Plaine CP231, Bruxelles, Belgium

3) Departament de Fisica i Enginyeria Nuclear, Universitat Politecnica de Catalunya, 08222 Terrassa, Spain
Gregory.Kozyreff@icfo.es , luis.domingues@icfo.es and jordi.martorell@icfo.es

Abstract: *We study theoretically surface second harmonic generation in microsphere using whispering gallery modes. Coupled-mode theory is derived. Conditions for double-resonance and phase matching are discussed. An experimental realization using a silica microsphere pumped with a tapered fiber is considered.*

Introduction

Since the demonstration that with the use of spherical microresonators one could couple the evanescent field of a single photon with a single atom,[i] it became clear the relevance that such type of resonators could have for the detection of very small concentrations of un-marked chemical or biological compounds. The high sensitivity of devices based on such type of microresonators resides on the high value of the quality factor for the cavity ($Q{\sim}10^8$) and on an evanescent field that penetrates the surrounding medium.[ii] The presence of an external agent on the outside surface of the microcavity produces a change in the evanescent wave, which translates into a detectable change in the light that propagates within the sphere in a whispering gallery mode (WGM).

The measurable changes come either from a change in the mass of the region where the evanescent wave propagates or from a change in the refractive index. To date, such microresonators have been used as biochemical sensors, where a change in the position of the resonance could be attributed to a change in the concentration of glucose,[iii] to quantify unmarked DNA,[iv] to detect the presence of certain proteins after a displacement of the resonance,[v] and to detect the presence of highly volatile organic compounds such as, for instance, toluene or nitrobenzene.[vi]

The performance of such micro resonators as sensors could be enhanced if the molecule to be detected played a direct role in the light generation process, or in a less favorable case, if the species to detect sat very close to the material that was used to generate the light.

In a process such as surface second harmonic generation in total internal reflection geometry, not only the propagation but also the light generation within the micro-resonator is directly related to the chemical composition of the surface. Although to reach a non-vanishing second order nonlinear process there are very strict requirements one must set on the parameters of the sphere, it has been proven, very recently, that it is possible to fabricate micro resonators of nonlinear material where the quadratic nonlinear interaction is efficient.[vii] However, the challenge to experimentally demonstrate a second harmonic interaction coupling the evanescent field with the radiation from a nonlinear dipole on the surface of the sphere remains.

Second Harmonic Generation in the WGM

We consider second harmonic generation in the WGM of a dielectric microsphere. On the surface of the sphere, inversion symmetry is broken, allowing the existence of a local second-order susceptibility.

We thus analyze the generation due to a surface nonlinearity.

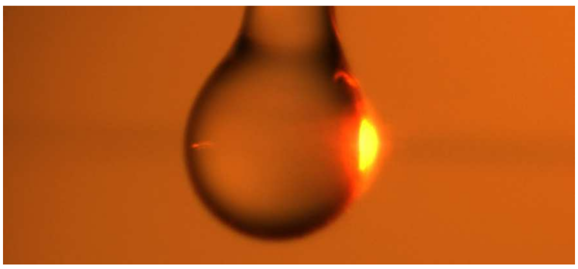

Fig. 1: Signature of fundamental WGM excitation. As the pump tapered fiber (shadow in the background) touches the sphere, an intense field is suddenly radiated at the rim of the sphere. Sphere diameter: 113 µm; wavelength: 0.8 µm.

A natural way to treat second harmonic generation is through a coupled-mode theory. We assume the electric field to be essentially described by two

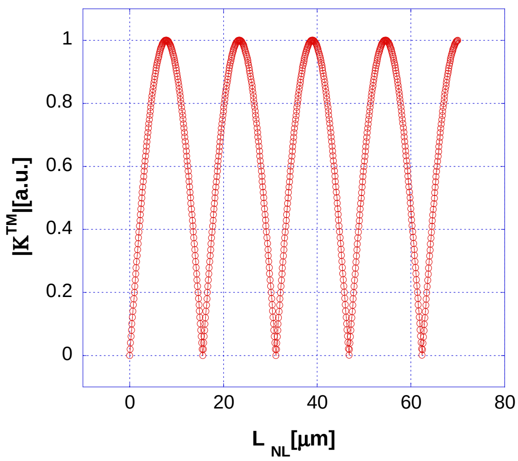

Fig. 2: Absolute value of nonlinear coupling, $|K^{TM}|$ as a function of the nonlinear coating length along the equator ($\mathbf{L_{NL}}$). Sphere radius R= 83 µm.

resonant modes of the sphere as

$$\mathbf{E} = \alpha_1(t)\mathbf{E}_{\omega_1}(\mathbf{r})e^{-i\omega_1 t} + \alpha_2(t)\mathbf{E}_{\omega_2}(\mathbf{r})e^{-i\omega_2 t} + c.c.,$$

where ω_1, ω_2 are resonant frequencies such that ω_1 is close to the pump frequency and $\omega_2 \approx 2\omega_1$. The resonant modes of the sphere $\mathbf{E}_{\omega_{j1}}(\mathbf{r})$ are given by the vector spherical harmonic solutions of Maxwell's equation. When the orbital number ℓ and the azimutal number *m* of such a solution are both large, this solution describes a Whispering Gallery Mode (WGM) of the cavity.

Such modes are typically excited by pumping light in the cavity with a tapered optical fiber, as shown in Figure 1. For a pump wavelength around 800 *nm* and a sphere radius larger than 30 *µm*, we have $\ell >> 1$ and the resonant frequencies are accurately given by asymptotic formulas[viii] . It is important to note that the spherical modes, being spherical waves away from the resonator, have an infinite energy (just as plane waves do). The amplitudes $\alpha_1(t)$ and $\alpha_2(t)$ are therefore normalized such that $|\alpha_j(t)|^2$ be the power (in watts) radiated by the j^{th} mode. Bearing this normalization in mind, we thus seek to derive a set of coupled-mode equations of the form

$$\frac{d\alpha_1}{dt} + \Gamma_1\alpha_1 = \Gamma_1\sqrt{P}e^{-i\Delta_1 t} + i\kappa^*\alpha_2\alpha_1^* e^{i(\Delta_2 - 2\Delta_1)t},$$

$$\frac{d\alpha_2}{dt} + \Gamma_2\alpha_2 = i\kappa\alpha_1^2 e^{-i\Delta_2 t}.$$

In these equations, P is the power that would be radiated by the fundamental mode in the absence of nonlinear conversion, taking into account the injection efficiency and the various losses suffered by that mode. On the other hand Δ_1, Δ_2 are the detuning between the injection frequency and the fundamental mode and the detuning between fundamental and second harmonic, respectively. Finally, Γ_j are the amplitude decay rates of the WGM. These are not only due to radiation, but also absorption and scatterin by surface impuritites.[ix] The purpose here is to derive the coupling strength κ between the two modes. Note that TM and TE WGM have very distinct electric field distribution in this limit and we must consider both cases separately.

Coupling strength and phase matching

The usual ray picture of a WGM suggests to understand phase matching in terms of two nearly-plane waves circulating along the equator of the sphere. This is however misleading. Indeed, WGM are cavity modes, i.e. they are confined to a closed geometry, which is quite distinct from free propagation. To demonstrate this point, we derive a coupled-mode theory from Maxwell's equation. We thus show that the coupling coefficient between a TM fundamental mode and a TM second harmonic with quantum numbers L and M obeys

$$\kappa^{TM} \propto \iint \chi^{(2)}_{s,\perp\perp\perp} Y^*_{LM} Y^2_{\ell m} \sin\theta d\theta d\varphi .$$

In this expression, $\chi^{(2)}_{s,\perp\perp\perp}$ is the appropriate component of the nonlinear susceptibility tensor. For a uniform nonlinear susceptibility, this integral will vanishes unless the quantum mechanical rules for angular momentum composition are satisfied. In a spherical cavity, therefore, phase matching corresponds to

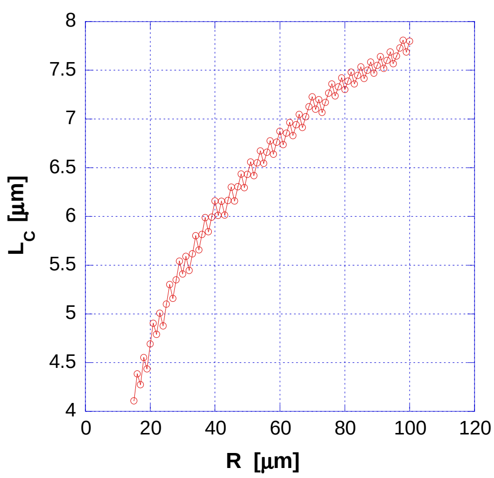

Fig. 3: Coherent Length ($\mathbf{L_C}$) for Second Harmonic Generation as function of the radio (**R**) of sphere.

conservation of angular momentum, instead of linear momentum for plane waves.

Furthermore, we show that TE fundamental modes are much more efficient to generate second harmonics. Indeed, we find that

$$\left|\kappa^{TE}\right| \sim \ell^2 \left|\kappa^{TM}\right|.$$

Except under the special favourable circunstance discussed above, the integral in κ^{TM} will vanish. A quasi-phase matching strategy is, therefore, to cover the surface of the sphere only over a distance L_{NL} along the equator. To determinate the coherent length of the sphere, let us suppose that:

$$\chi^{(2)}_{s\perp\perp\perp} = \begin{cases} \chi^{(2)}_s & \text{for} \quad 0 < \varphi < L_{NL}/R, \\ 0 & \text{otherwise,} \end{cases}$$

R being the radius of the sphere and φ the azimutal angle. One obtains the periodic dependence $\kappa^{TM}(L_{NL})$ depicted in Figure 2. The coherence lengh L_C is thus defined as half the speration between two maxima of $\left|\kappa^{TM}\right|$.[x] In Figure 3, we plot the coherent length L_C as a function of R. Note that the coherence length is not constant as the sphere radius varies, due to the index modal dispersion of the WGM. The discontinuous character of the curve results from jumps in the values of ℓ and L corresponding to the most resonant fundamental and second harmonic modes as R is varied. The same approach can of course be followed for more realistic nonlinear coatings, e.g. spherical caps of varying diameter L_{NL}.

Conclusion

We have discussed the possibility of second harmonic generation (SHG) in a microsphere due to a surface nonlinearity. This system, apart from fundamental interest, has promising applications in sensor technology. To describe SHG, a coupled-mode theory based on Whispering Gallery Modes is derived, for which the main result is an analytical expression for the coupling strength between fundamental and second harmonic modes. Phase- and quasi-phase matching is still possible in this system, but as the expression for the coupling constant clearly indicates, frequency mixing now requires conservation of angular momentum. Furthermore, nonlinear generation is ℓ^2 times more efficient with a TE fundamental mode than with a TM fundamental mode. Finally, the concept of coherence length was revisited and discussed for the spherical geometry.

Silica microspheres of the adequate diameter have been prepared to demonstrate such predictions of the coupled-mode theory. We have shown that it is possible to cover the surface of the sphere with a layer of nonlinear material. We also present the realization of the experimental setup to measure second harmonic generation in the Whispering Gallery Modes of the spherical resonator.

References

[i] D. W. Vernooy et al. Phys. Rev. A, Vol. **57**, R2293 (1998).

[ii] K. J. Vahala, "Optical Microcavities", *Nature*, Vol. **424**, pp. 839-846, 2003.

[iii] Chung-Yen Chao and L. Jay Guo, Appl. Phys. Lett., Vol. **83**, 1527 (2003).

[iv] F. Vollmer et al., Biophysical Journal, Vol. **85**, 1974 (2003).

[v] F. Vollmer et al., Appl. Phys. Lett., Vol. **80**, 4057 (2002).

[vi] W. Fang et al. Appl. Phys. Lett., Vol. **85**, 3666 (2004).

[vii] V. S. Ilchenko et al., Phys. Rev. Lett., Vol. **92**, 043903 (2004).

[viii] C.C. Lang, P.T. Leung, and K. Young, J. Opt. Soc. Am. B, Vol. **9**, 1585 (1992).

[ix] M.L. Gorodetsky, A.A. Savchenkov, and V.S. Ilchenko, Opt. Lett., Vol. **21**, 453 (1996).

[x] M. M. Fejer, G. A. Magel, D. H. Jundt, and R. L. Byer. IEEE J. Quantum Electron., Vol. **28,** 2631–2654 (1992).