On the fundamental limits of Q factor of crystalline dielectric resonators

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Abstract: The temperature dependence of the processes which fundamentally limit optical quality factor of ideal crystalline whispering gallery mode resonators is investigated. The example of CaF_2 is used to show that spontaneous Raman scattering is the main limitation of the quality factor at low temperatures. Stimulated Raman scattering is also shown to be important at any temperature. We experimentally demonstrate nonlinear absorption due to stimulated Raman scattering in a real cavity at room temperature and theoretically derive Raman gain of CaF_2. We conclude that optical storage times in excess of one second could be achieved in millimeter sized cavities.

OCIS codes: (190.0190) Nonlinear Optics, (290.0290) Scattering, (230.5750) Resonators.

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1. Introduction

Recent progress in fabrication of whispering gallery mode (WGM) resonators with crystalline materials has resulted in the highest demonstrated quality factors (Q) for this type of cavities [1, 2]. The mechanical polishing techniques used in the fabrication process essentially eliminate surface scattering and surface absorption as loss mechanisms at the achieved material transparency levels [3]. The only remaining physical mechanisms limiting the ultimate achievable Q with this approach are linear and nonlinear attenuation in the dielectric material.

Optical transparency of ideal dielectric crystals is restricted by the fundamental blue and red wings of photon absorption. The blue wing is defined by optically allowed transitions between valence and conduction electron bands. The red wing results from the interaction between light and phonons in the crystalline medium. Both absorption mechanisms have a strong frequency dependence, so the absorption of light in a dielectric within the transparency band can be extremely low. The question is how low it can be, and is there any other transparency limiting mechanism. Similar question has been discussed for optical fibers in Ref. [4]. It was shown that the scattering loss mechanisms such as Rayleigh, Brillouin and Raman determine the extent of optical attenuation. These mechanisms are also present in ideal optical crystals and limit their transparency as well.

In this paper we analyze the minimum light attenuation achievable with ideal crystals at room and cryogenic temperatures. We use a real almost perfect fluorite crystal as an example. It was shown previously that the attenuation of UV light in artificially grown CaF_2 is nearly completely limited by Brillouin and thermodynamic Rayleigh scattering [5] at room temperature. We show that the spontaneous Raman scattering is primarily responsible for the attenuation in an ideal crystal at low temperatures. Applying this result to the crystalline WGM resonators we find the limits to the IR quality factors. We also show that, in contrast to the low temperature regime, spontaneous Brillouin scattering as well as stimulated Raman scattering (SRS) determine the Q factor of a WGM resonator under realistic experimental conditions at room temperature. We measure the influence of SRS on the Q factor of a millimeter sized resonator using a ring-down technique and show that this scattering becomes important at input power levels as low as a few microwatt.

2. Three types of scattering

Let us consider Rayleigh, Brillouin and Raman scattering in a perfect crystal. Thermodynamically limited Rayleigh scattering is small in an ideal crystal, and Brillouin scattering dominates. The quantitative ratio of Rayleigh and Brillouin scattering can be estimated using the Landau-Plachek relationship (see, e.g., Ref. [6]):

$$\frac{\alpha_{Ri}}{\alpha_B} \simeq \frac{\beta_T}{\beta_S} - 1 \tag{1}$$

where β_T and β_S are the isothermal and isobaric compressibilities, respectively. This ratio is generally less than unity in crystals and vanishes with temperature decrease in the low temperature limit (Ref. [7], p. 219):

$$\frac{\beta_T}{\beta_S} - 1 = \frac{C_P}{C_V} - 1 \sim T^4|_{T\to 0}, \tag{2}$$

where C_P and C_V are the specific heat capacities at constant pressure and constant volume, respectively. Rayleigh scattering can be neglected in a perfect crystal at low temperature, as the result of (2).

Temperature dependence of spontaneous Raman and Brillouin scattering mechanisms is determined by the number of participating phonons, given by a Bose population factor [8]. The

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corresponding light attenuation coefficient accounting for both Stokes and anti-Stokes components may be estimated as follows (see also Ref. [6], p. 99)

$$\alpha_{B,R} \approx \alpha_{B0,R0} \left(\frac{\lambda_0}{\lambda}\right)^4 \left[\left(\exp\frac{\hbar\Omega_{B,R}}{k_B T} - 1\right)^{-1} + \frac{1}{2}\right] \tag{3}$$

where λ is the wavelength of light, $\alpha_{B0,R0}$ are the scattering parameters given by the properties of a particular crystal corresponding to λ_0, Ω_B is the Brillouin frequency shift for 90^o scattering and Ω_R is the Raman frequency shift.

In writing of Eq. (3) we have assumed that the crystal has a single phonon branch. Using data evaluated for CaF_2 at $T = 300$ K in Ref. [5]: $\hbar\Omega_B/k_B T \simeq 1/300$, $\hbar\Omega_R/k_B T \simeq 1/4$, $\lambda_0 = 0.532\ \mu m$, $\alpha_B = 2.411 \times 10^{-7}$ cm^{-1}, $\alpha_R \simeq 4.34 \times 10^{-8}\ cm^{-1}$ we obtain $\alpha_{B0} \simeq 8 \times 10^{-10}$ cm^{-1}, $\alpha_{R0} \simeq 1.1 \times 10^{-8}\ cm^{-1}$. It is easy to see that Brillouin scattering is significantly suppressed and the attenuation of light in a perfect crystal is determined by the spontaneous Raman scattering at low temperature. The room temperature attenuation, on the other hand, is given by spontaneous Brillouin scattering.

It is shown in Ref. [5] that the existing calcium fluoride crystals have transparency close to the fundamental limit (3) in the UV. However, such a low attenuation is yet to be demonstrated in experiments with visible and infrared light. The measured transparency is two to three orders of magnitude lower than the fundamental limit in those frequency bands [1, 2, 3] because of the extrinsic and intrinsic impurities of the material.

3. Properties of fluorite WGM resonators

We now discuss the optical properties of fluorite WGM resonators. The maximum quality factor of the resonator is given by $Q_{max} = 2\pi n(\lambda)/[\lambda\alpha(\lambda)]$, where $n(\lambda)$ is the refractive index of the material, λ is the wavelength of light in vacuum, and $\alpha(\lambda)$ is the total loss coefficient of the bulk dielectric material. This formula is not entirely correct for scattering losses. A coherent scattering loss, where some part of scattered light is deflected back into the mode, leads to the enhancement of the Q factor by up to an order of magnitude [9]. We still use the formula as the role of scattering is minor in the case of an ideal fluorite cavity, which we consider in this study. In such a cavity the surface roughness is given by the lattice constant and the corresponding optical storage time can reach 1s for a resonator having 1cm in diameters for $\lambda = 1\mu m$.

In case of a real cavity, the surface roughness can limit Q factor. Some estimates made in Ref. [2] for a particular fluorite cavity show upper limit for Q of about 10^{12}. However, this is a technical limitation given by the particular polishing procedure. A further investigation is required before storage time beyond 1 second is achieved.

In what follows we neglect the surface scattering and consider only unavoidable attenuation mechanisms. The corresponding wavelength dependence of the attenuation as well as Q factor for CaF_2, given by Raman and Brillouin scattering as well as by blue and red wing absorption are shown in Fig. 1. The approximation of the maximum quality factor of CaF_2 WGM resonators found in Ref. [1] using existing experimental results nearly coincides with the fundamental limit presented in this paper at room temperature. The wavelength dependence of the index of refraction of the material at $T = 300$ K was found using the 4-term Sellmeier equation [10]. Blue and red absorption wings can generally be approximated by a simple exponential dependence [11, 12]. Parameters for the blue wing have been derived from Ref. [13]. Red wing parameters were obtained from Ref. [14] and the experimental data publicly available for Corning CaF_2. For our analysis we have assumed both wings to be temperature independent, given that the real temperature dependence is weak. According to Fig. 1, the lifetime of a photon in a fluorite WGM resonator can exceed 1 second for $\lambda = 1.5\mu m$.

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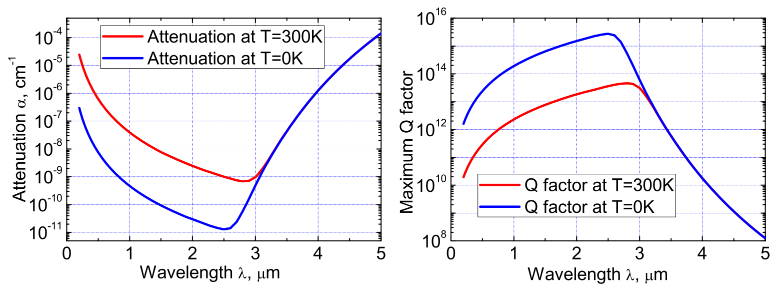

Fig. 1. Attenuation in ideal CaF_2 (left) and Q factor (right) of an ideal fluorite WGM resonators at room and nearly absolute zero temperature. Contributions from spontaneous Brillouin, Rayleigh and Raman scattering as well as blue and red wing absorption are added.

We now show that not only spontaneous, but also stimulated scattering is important in macroscopic WGM resonators. Stimulated Rayleigh scattering is not experimentally observable in high Q WGM resonators as it is suppressed by a mismatch of modal structure of a typical cavity and the gain profile for this process [6]. Stimulated Brillouin scattering is suppressed because of the absence of phase matching between acoustical and optical modes in the WGM resonators. The resonantly enhanced stimulated Raman scattering is always possible if the frequency difference between subsequent modes of the same mode family (free spectral range, FSR) is less than the spectral width Γ of the optical phonons. In fluorite at room temperature $\Gamma = 450$ GHz [15]. Hence any resonator with radius $R \geq c/(2\pi n(\lambda)\Gamma) \simeq 75\ \mu$m is not immune to SRS.

Stimulated Raman scattering, as well as spontaneous Raman scattering, is present in even an ideal crystalline lattice at nearly absolute zero temperature. It is one of the fundamental causes of light absorption in a WGM resonator. The stimulated scattering generates optical phonons in the material. The phonons decay due to the multiphonon scattering, which results in heating of the crystal and growing of the entropy. To estimate the maximum quality factor achievable in a crystalline WGM resonator if SRS is allowed we consider a resonator supporting a pump and several first order Stokes modes. Such a system can be described at low temperature by a set of rate equations [16]:

$$\dot{n}_S = -2\gamma_S n_S + \hbar\omega_p \frac{c^2}{n^2}\frac{g_b}{V} n_p n_S, \tag{4}$$

$$\dot{n}_p = -2\gamma_p n_p - \hbar\omega_p \frac{c^2}{n^2}\frac{g_b}{V} \sum_S n_p n_S, \tag{5}$$

where n is the refractive index, $n_p \gg 1$ and $n_S \gg 1$ are the averaged photon numbers in the pump and Stokes modes, V is the mode volume and $2\gamma_S = \alpha(\lambda_s)c/n$ and $2\gamma_p = \alpha(\lambda_p)c/n$ are the total linear decay rates of Stokes and pump modes respectively. The rates take spontaneous scattering decay into account. We assume that the modes completely overlap and have the same volume V. These equations were derived from (25) and (26) of Ref. [17] by replacing spatial derivatives ($dP_{p,S}/dz$, $P_{p,S}$ is the power of pump and Stokes light) with temporal derivatives ($\dot{n}_{p,S}$).

The bulk Raman gain g_b is related to the attenuation due to spontaneous Raman scattering as

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(Ref. [16], p.371)

$$g_b(\lambda_p) = \frac{\alpha_R(\lambda_p)\lambda_p^4}{8\pi^2 c^2 n^2}\frac{\omega_p}{\hbar\Gamma}, \tag{6}$$

where we assumed that $\lambda_s = \lambda_p$. The wavelength dependence of the gain is shown in Fig. 2C. It is easy to see that for CaF_2 the value of the bulk Raman gain is $g_b = 2.4 \times 10^{-11}$ cm/W at $\lambda = 1\mu$m.

An approximate temperature dependence of the spectral width of the Raman phonons is given by [18]

$$\Gamma = \Gamma_0 \left[1 + \frac{2}{\exp(\hbar\Omega_R/2kT) - 1}\right] \tag{7}$$

if we assume that phonons experience degenerate two–particle anharmonic decay.

According to Eq. (5) the effective decay rate of the pump radiation due to SRS is

$$2\gamma_{SRS} = \hbar\omega_p \frac{c^2}{n^2}\frac{g_b}{V} N_S n_S, \tag{8}$$

where N_S is the total number of Stokes modes the light decays into, and n_S depends on the pump power. The quality factor of the pump mode, Q_{SRS}, is going to be smaller than Q when the pump power exceeds P_{th} [19]:

$$\frac{Q}{Q_{SRS}} = \sqrt{\frac{P}{P_{th}}}, \tag{9}$$

where SRS threshold power is given by

$$P_{th} = \frac{\pi^2 n^2}{g_b Q_p Q_S}\frac{V}{\lambda_p \lambda_S}, \tag{10}$$

and the external pumping P is assumed to exceed the threshold significantly $P \gg P_{th}$. Selecting the realistic values for a 1mm cavity $g_b = 2.4 \times 10^{-11}$ cm/W, $n = 1.43$, $V = 1.4 \times 10^{-7}$ cm^3, $Q_p = Q_S = 10^{10}$, $\lambda_p = \lambda_S = 1\ \mu$m, we obtain $P_{th} \simeq 120$ nW. Pumping the resonator with 1μW of light for these parameters will result in a reduction of the effective quality factor of the corresponding WGM down to $Q_{SRS} = 3.5 \times 10^9$. It should be emphasized that this quality factor is related to the pump light only. To observe this Q factor decrease, the Stokes light should be blocked from reaching the photodetector when using a ringdown technique. We may combine Eq. (10) with Eqs. (7) and (6) to derive temperature dependence of SRS threshold power for this cavity. It is also interesting to estimate the threshold power in terms of a photon number given by the expression $P_{th}T_R/\hbar\omega$, where $T_R = Q/\omega$ is the ringdown time. The results are presented in Fig. 2.

4. Experimental observation of nonlinear attenuation

We have performed an experiment to demonstrate the nonlinear attenuation caused by SRS. Measurements at 780 nm were performed with a diode laser. Free beam emission from the laser was coupled to a fiber, which had an output collimator attached. Light from the collimator lens was sent into a glass prism, which served as a coupler for a CaF_2 WGM resonator. Coupling efficiency of the pump light to WGMs was better than 30%. The output light that went through the coupler was collimated with a lens and collected on a photodetector. Although the Stokes light should be blocked to observe the nonlinear behavior, in this experiment we did not employ specific measures to do so. We suppose the Stokes light was partially blocked accidentally by dispersive properties of the setup components.

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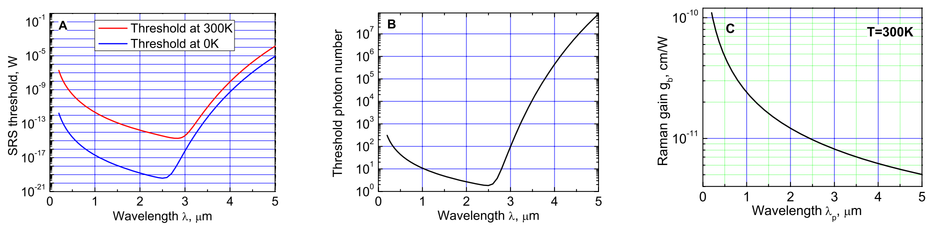

Fig. 2. A) SRS threshold for a 1 mm ideal-surface cavity made with an ideal CaF_2. B) Low temperature SRS threshold for a 1 mm ideal cavity made with CaF_2 in terms of photon number. C) Theoretically evaluated wavelength dependence of the Raman gain in CaF_2.

We observed ringdown signals obtained by periodic scanning of the laser frequency over many resonator modes with the internal piezo of the laser head. Time–dependent signals resulted from the interference between the pump radiation and the emission from the resonator. According to our observations the signal had a clear nonexponential time dependence (Fig. 3), which supports the above discussion.

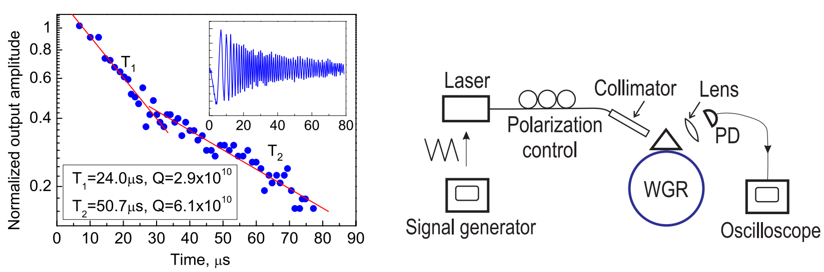

Fig. 3. Experimental observation of the ringdown signal of a calcium fluoride resonator and setup scheme. The observations were performed at 780 nm. The threshold power of the SRS process was on the order of a few microwatt. Linear fits of the amplitude decay are presented by red lines, emphasizing two different ringdown constants at the beginning and at the end of the process. Inset shows the actual signal recorded on the photodetector. Blue points in the graph represent the amplitude of the signal shown in the inset.

5. Conclusion

We have presented theoretical as well as experimental studies of light attenuation resulting from fundamental scattering in crystalline WGM resonators. We show that the spontaneous Raman scattering results in the fundamental restrictions of the resonator quality factor at low temperatures. Stimulated Raman scattering also restricts the quality factor at any temperature if the power of the light used in the resonator exceeds the threshold of the stimulated process.

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