Millisecond Photon Lifetime in a Slow-Light Microcavity

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Optical microcavities with ultralong photon storage times are of central importance for integrated nanophotonics. To date, record quality (Q) factors up to 10^{11} have been measured in millimetric-size single-crystal whispering-gallery-mode (WGM) resonators, and 10^{10} in silica or glass microresonators. We show that, by introducing slow-light effects in an active WGM microresonator, it is possible to enhance the photon lifetime by several orders of magnitude, thus circumventing both fabrication imperfections and residual absorption. The slow-light effect is obtained from coherent population oscillations in an erbium-doped fluoride glass microsphere, producing strong dispersion of the WGM (group index $n_g \sim 10^6$). As a result, a photon lifetime up to 2.5 ms at room temperature has been measured, corresponding to a Q factor of 3×10^{12} at 1530 nm. This system could yield a new type of optical memory microarray with ultralong storage times.

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Optical microresonators with very long cavity lifetimes and very high quality (Q) factors are of great interest in contemporary photonics [1,2]. In the linear regime, ultrahigh-Q-factor microresonators can be used as optical memories [2], delay lines, or highly selective filters for optical [3–5] or microwave photonics applications [6,7], bio- or chemical optical sensors [8]. In all cases their miniaturized and monolithic feature leads to a high stability and integration potential. Several microcavity topologies are currently studied; among them, whispering-gallery-mode (WGM) resonators have received a lot of attention because they allow the highest photon lifetimes to be obtained [9,10]. The Q factor (Q) and the photon lifetime (τ_{cav}) of a microcavity resonant for an angular frequency $\omega_0 = 2\pi c/\lambda_0$ considering a Lorentzian profile (see the details in the Supplemental Material [11]) can be written

$$Q = \omega_0 \tau_{\mathrm{cav}} = \frac{n_g(\omega_0) L \mathcal{F}}{\lambda_0}, \tag{1}$$

where n_g is the group index of the material constituting the microcavity of finesse $\mathcal{F}$ and L is the cavity round-trip length. To reach ultrahigh Q factors ($\simeq 10^{10}$), research groups usually focus their efforts on increasing the finesse value by reducing optical losses due to absorption or scattering [9,16–19]. Until now, the longest photon storage times have been obtained using WGM miniresonators made from highly transparent materials such as fluoride crystals [20]. Thanks to a careful polishing at the limit of conventional techniques and to an optimized annealing procedure, Q factors up to 10^{11} have been measured in a crystalline CaF_2 resonator [21] corresponding to a record photon storage time of 100 μs at $\lambda_0 \approx 1550$ nm. Such high Q factors are reached by using resonators with a diameter of a few millimeters, which reduces the detrimental role of the surface scattering and Rayleigh, Brillouin, or Raman scattering [20].

Another way to significantly increase the photon storage time, and thus the Q factor, consists in introducing a strongly dispersive medium in the cavity [22,23]. Equation (1) shows that, for a given finesse, the cavity Q factor can be strongly enhanced using a dispersive or slow-light medium characterized by a high group index. Few demonstrations of this effect have been done in macroscopic optical cavities using atomic beam or vapors [24,25]. The main idea is to use resonant coherent effects such as electromagnetic induced transparency (EIT) or coherent population trapping (CPT) to achieve simultaneously a low absorption to preserve the finesse of the cavity and a strong dispersion [23]. The steep refractive index variation due to the interaction between a "high" intensity pump and a probe fields via the atomic medium leads to a low group velocity which artificially increases the optical path of the probe, and thus the photon storage time. For solid-state cavities, coherent or nonlinear effects can also be used to produce the linewidth narrowing. Recently, a 4 orders of magnitude Q-factor enhancement (from 5×10^5 to 1.7×10^{10}) has been observed using persistent hole burning in a 6 mm-long praseodymium doped Y_2SiO_5 cavity [26].

In 2005, Soljačić *et al.* [27] have theoretically demonstrated that this method could be used to significantly enhance the lifetime of microcavities, for which the limiting factors are the residual absorption of the constitutive

material and the leakage towards undesired modes. They have predicted that the dispersion contributes in improving all of the Q factors associated with absorption and the radiative loss, enhancing thus the overall Q factor of the microcavities. More recently, in the first paper reporting the experimental demonstration of slow-light cavity lifetime increase at solid state, Sabooni *et al.* [26] proposed that the extension of their work to rare earth doped WGM microcavities would be of great interest for obtaining long photon lifetime microresonators. This method, which is radically different from finesse improvement techniques, can circumvent the limitations of surface scattering in passive resonators [20] or internal gain saturation in active microresonators [18], leading to ultrahigh-Q-factor micron-size resonators.

In this Letter we report the implementation of these propositions and show that it is also possible to reach the regime of ultraslow light in optical microcavities. We have used an emblematic WGM cavity: a spherical glass microcavity doped with erbium ions. Relying on coherent population oscillations (CPOs), this approach, which is easily transposable to other materials or coherent effects, is used to overcome optical and technological losses and achieve unprecedented millisecond photonic lifetimes and associated 10^{12} Q factors in microresonators at room temperature.

Figure 1 is a sketch of a three-port erbium-doped ZBLALiP fluoride glass [28] WGM microsphere resonator. Unlike EIT, which requires a particular three-level Λ atomic system, CPO is a coherent effect which can be observed at room temperature in any two-level system (TLS) [29–31] and is suitable for the demonstration of fundamental phenomena using slow light [32]. In our experiment, the TLS required to implement the CPO effect is obtained using the ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of the Er^{3+} ion. The spheres were obtained by the fusion of glass powders using a microwave plasma [18]. We assume that the pump field

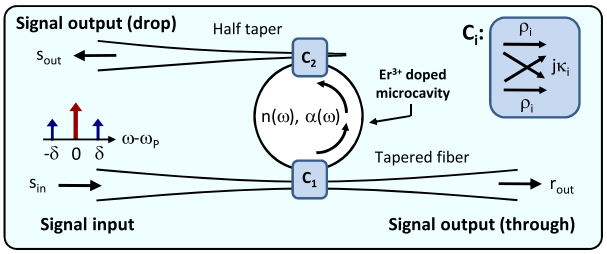

FIG. 1. Schematic view of a three-port WGM erbium-doped microsphere. The input signal s_{in} is injected within the microcavity using a tapered fiber which can also be used to collect the output signal r_{out} on the through port. A half tapered fiber is used to extract the drop port output field (s_{out}). The coupling between the microsphere (characterized by its absorption α and refractive index n) and the tapers is modeled using transfer matrices with repartition coefficients ρ_i and $j\kappa_i$, with $i \in \{1, 2\}$ and $(\rho_i, \kappa_i) \in \mathbb{R}^2$, such that $\rho_i^2 + \kappa_i^2 = 1$.

$s_{in}(\omega_P)$ angular frequency is tuned to the cavity resonance ($\omega_P = \omega_0$). We consider that the pump wavelength is $\lambda_P \approx 1530$ nm corresponding to the absorption maximum of the Er^{3+} ion in ZBLALiP [33]. The Q-factor enhancement is probed using two sidebands $s_{in}(\omega_P \pm \delta)$. In the limit of a short dephasing time T_2 compared to the excited state lifetime T_1 ($T_2 \ll T_1$) and assuming that the pump field is tuned to the atomic resonance, for $\delta \neq 0$, the absorption α and the refractive index n at the probe frequency ($\omega_P + \delta$) are given by (see Ref. [11])

$$\alpha(\omega_P + \delta) = \frac{\alpha_0}{1+S}\left(1 - \frac{2S(1+S)}{(1+S)^2 + (\delta T_1)^2}\right)$$
$$n(\omega_P + \delta) = 1 + \frac{c\alpha_0}{\omega_P(1+S)}\frac{S\delta T_1}{(1+S)^2 + (\delta T_1)^2}, \quad (2)$$

where S is the intracavity pump intensity normalized by the saturation intensity and α_0 the unsaturated absorption varying between 0.09 and 0.11 cm^{-1}, depending on the Er^{3+} ion density and pump wavelength. The first equation shows that, for a probe such as $\delta T_1 \ll 1$, a narrow transparency window is created in the absorption profile of the TLS. This absorption dip is associated with a strong refractive index variation characterized by the group index $n_g = n + \omega(dn/d\omega)$:

$$n_g(\omega_P + \delta) \approx \frac{\alpha_0 c T_1 S}{(1+S)^3}. \quad (3)$$

The Er^{3+} ion is attractive for slow light because of its long lifetime, $T_1 \approx 10$ ms, leading to a huge group index ($n_g > 10^6$). In our case, this property will be used to strongly enhance the WGM resonator photon storage time, as shown in Eq. (1).

We first performed experiments to demonstrate the ultraslow-light propagation regime in our WGM microspheres. In bulk materials, this is usually done by measuring the group delay τ_g introduced by the slow-light medium [29,34]. For the cavity configuration shown in Fig. 1, assuming a strongly coupled cavity such as $\kappa_1^2 + \kappa_2^2 \gg \alpha_0 L/8$ to avoid absorption bistability and neglecting its nonlinear behavior [35,36], the probe group delay for $\delta T_1 \ll 1$ is given by (see Ref. [11])

$$\tau_g(\omega_P + \delta) \approx \frac{n_g(\omega_P + \delta)L}{c} \times \frac{\mathcal{F}}{\pi}. \quad (4)$$

Thus, the overall group delay reaches the value that it would be obtained in a slow-light medium with an effective length $L \times \mathcal{F}/\pi$. To measure the group delay, a single-mode pump laser at $\lambda_P = 1530$ nm is intensity modulated with a $1 + m\cos(\delta t)$ wave function using an acousto-optic modulator (AOM) (see the experimental setup sketch in Ref. [11]). For low values of m (here, $2m = 10\%$), the input

signal is thus constituted by the pump at ω_P and two weak sidebands at $\omega_P \pm \delta$ acting as the probes. This input signal is injected via a tapered fiber in the WGM microsphere and the output signal is detected on the drop port using a half tapered optical fiber. We deduce the group delay from the phase shift $\Phi = \Phi_s - \Phi_{\mathrm{ref}}$ between the input modulation (Φ_{ref}) and the output modulation (Φ_s) by measuring $t_g = \Phi/\delta$. The relation between Φ and the refractive index can be found in Ref. [11]. Since $\omega_P(dn/d\omega_P)|_{\omega_P} \gg 1$, it can be shown that

$$t_g(\delta) = \frac{\omega_P L\mathcal{F}}{\pi c}\frac{n(\omega_P+\delta)-n(\omega_P-\delta)}{2\delta} = \frac{\alpha_0 T_1 L\mathcal{F}}{\pi}\frac{S}{1+S}\frac{1}{(1+S)^2+(\delta T_1)^2}. \quad (5)$$

Consequently, for $\delta T_1 \ll 1$ we have $t_g(\delta) \approx \tau_g(\omega_P+\delta)$, and thus t_g is a good estimation of τ_g. For this experiment we used a 0.1 mol% erbium-doped microsphere with a diameter $D = 95\ \mu$m. First, we selected (without the intensity modulation on) a WGM resonance near the absorption maximum (1530 nm) by continuously sweeping the pump wavelength over 9 GHz. After a calibration of the frequency sweeping rate, we recorded the through and drop transmission spectra, as shown in the inset of Fig. 2. The through port is used to find and monitor the resonance, whereas the drop port is used to perform the group delay and cavity lifetime measurements. The width of the pump resonance is thus estimated to 600 MHz corresponding to a pump quality factor of $Q_P = 3.3 \times 10^5$ or a resonant group delay of 530 ps (see Ref. [11]). Then the pump laser is finely tuned to this resonance (ν_0) and modulated using the AOM. The measurement of $t_g(\delta)$ is reported in Fig. 2 for two different power values at the input of the tapered fiber $P_{\mathrm{in}} = -23$ dBm and $P_{\mathrm{in}} = -20$ dBm. The two experimental data sets are *simultaneously* fitted using S (the normalized intensity for $P_{\mathrm{in}} = -23$ dBm), T_1, and $\mathcal{F}$ as free parameters. We found $T_1 = 10.7$ ms, which is in good agreement with the measured lifetime (≈13 ms) of the level $^4I_{13/2}$ in ZBLALiP for the erbium density we used [28]. Note that we have found that the value of T_1 measured in our microspheres is around 20% lower than that measured in the bulk glass, as has been already reported for other materials [37]. The value of the finesse deduced from the measurements is $\mathcal{F} = 1100$, which gives a pump quality factor $\lambda_P\mathcal{F}/(\pi n_0 D) = 3.2 \times 10^5$ (where $n_0 = 1.5$ is the refractive index of ZBLALiP at ω_P), in very good agreement with the value deduced from the direct measurement. Additional experimental results on the probe transmission and the pump power dependence of the probe group delay are given in Ref. [11].

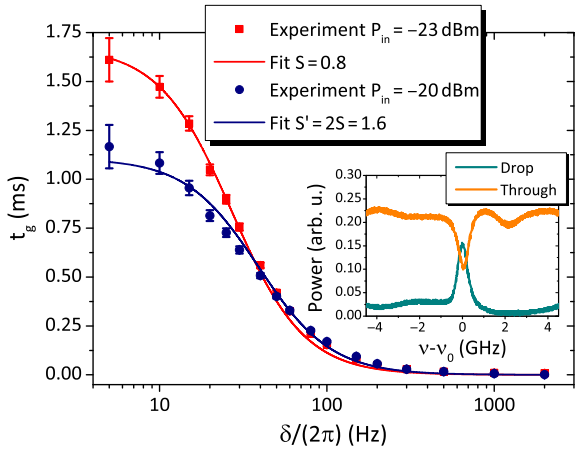

FIG. 2. Experimental probe group delay t_g for two input powers. The intracavity normalized power values S are deduced from the fit. We reduced the number of free parameters by simultaneously fitting the two sets of data using the supplementary constraint $S' = 2S$ for the saturation parameter associated with $P_{\mathrm{in}} = -20$ dBm. Note that $S = 0.8$ corresponds to a group index of 4×10^6. (Inset) Pump transmission spectra on the drop and through ports. $\nu = \omega/(2\pi)$ is the frequency of the tunable laser and ν_0 the resonance frequency of the cavity.

Beyond the resonant group delay, we also measured the photon lifetime of the hybrid cavity (i.e., the WGM resonator containing the slow-light medium) using a cavity ring-down technique. For this purpose, we change the waveform of the input signal: after a few oscillating cycles, the two sidebands are suddenly suppressed by turning off the modulation of the AOM ($m = 0$). The cavity lifetime is deduced from the fit of the exponential decay of the output signal modulation $M(\delta)$, which is proportional to the square root of the probe power transmission (see the details in Ref. [11]). The experimental results are given in Fig. 3(a) for two different experimental configurations. We also give the two signals detected on the reference channel to show that the exponential decay is not due to the instrument temporal response. The output signal has two exponential components. The first is related to the cold cavity and has a very short decay time of approximately 100 ps for a Q factor of around 10^5. Note that this exponential decay is not temporally resolved with our experimental setup (see the Supplemental Material [11] for details on the experimental setup). The second is very long and corresponds to the probe photons seeing the combined effect of the cavity and the CPO. For the two cases, we also give the group delay deduced from the modulation phase shift measurement. In Fig. 3(b) we report the theoretical calculations which have been carried out using a rate equation model [35,38–40] well suited for modeling the dynamical properties of nonlinear cavities [36,41] (see Ref. [11]). The only free parameter in the calculation is the input amplitude (s_0 or S). We adjusted this value to obtain in the stationary regime the corresponding values of t_g. We obtain a good agreement between theory and experiments for both the field lifetime (τ) and the amplitude of the decay signal. This experiment enables us to estimate twice the cavity lifetime since we

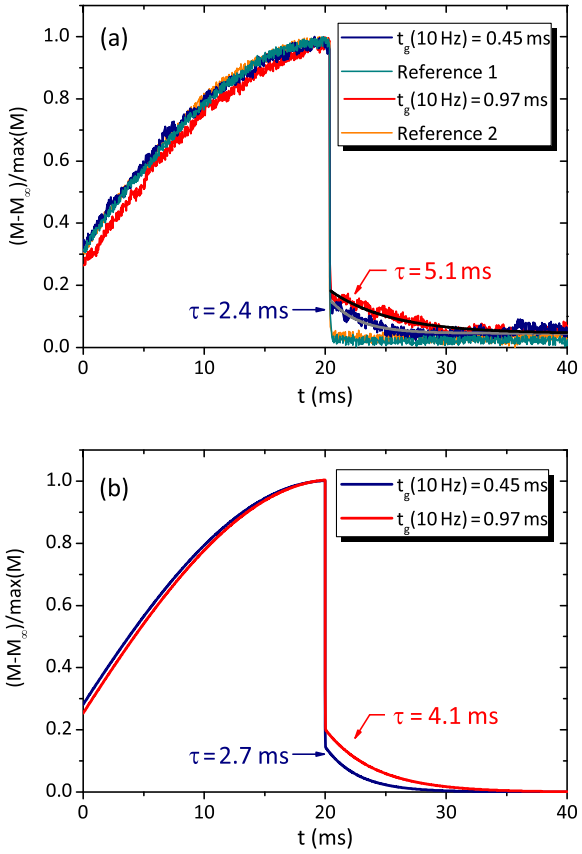

FIG. 3. The normalized modulation amplitude, M_∞, is the output modulation amplitude for $\delta T_1 \to \infty$. (a) Cavity ring-down measurements for two different experimental conditions. At $t = 20$ ms, the modulation is switched off. For the two experiments, we also give the signal detected on the reference photodiode. (b) Theoretical calculations obtained using the model described in Ref. [11]. The nominal values of excited lifetime state and pump wavelength are $T_1 = 10$ ms and $\lambda_P = 1530$ nm, respectively. For $t_g = 0.45$ ms, $Q_P = 1.8 \times 10^5$, $\tau_{a0} = 0.9$ ns, $S = 3.3$. For $t_g = 0.97$ ms, $Q_P = 2.6 \times 10^5$, $\tau_{a0} = 1.1$ ns, $S = 2.1$.

measure the beating between the pump and the probe [41]. For the optimized configuration giving $t_g = 0.97$ ms, we deduce a photon storage time for the probe signal $\tau_{\mathrm{cav}} = \tau/2 = 2.5$ ms.

We have shown experimental evidence for ultraslow-light propagation in a WGM microresonator with a group index around 4×10^6. The strong light slowing down enables us to increase the photon storage time of the cavity from 210 ps to 2.5 ms. The photon lifetime ($\tau_{\mathrm{cav}} = 2.5$ ms) of the hybrid cavity corresponds to a Q factor of 3×10^{12} which is 2 orders of magnitude larger than the Q factor of erbium-doped amplifying microspheres [18]. This measurement is in qualitative agreement with a value of 6×10^{11}, which can be estimated from the group delay measurement $t_g = 0.97$ ms and using results given in Ref. [11]. There are two reasons for the discrepancy between the two values: (i) the group delay is not measured for $\delta = 0$, and (ii) the cavity resonance does not have a Lorentzian profile, which substantially modifies the usual relation between the resonant group delay and the Q factor. Nevertheless, in the stationary regime, the experimental results are well reproduced by a simple model of dispersive medium embedded in a high finesse cavity. A numerical model has been used to study the dynamic behavior of the cavity and predict with good agreement the photon storage time. From an experimental point of view, the detrimental effect of the fast relaxation decay of the cold cavity, which reduces the amplitude of the decay signal in the cavity ring-down measurements (see Fig. 3), could be overcome by using the nonlinear behavior of the microresonator. Numerical simulations show that, in the undercoupling regime, the absorption bistability regime could be reached leading to a strong differential amplification [36], and thus to a Lorentzian shape for the probe resonance [35]. Unlike four wave mixing, the CPO effect is not sensitive to phase mismatch [39]. Consequently, a noncollinear pump and probe beam configuration would be interesting to spatially filter the pump and probe fields, which are slightly detuned. To reach such a high Q-factor value (3×10^{12}) using fluoride crystal millimetric diameter WGM resonators, the surface roughness would be around 0.23 nm (see the details of the calculations in Ref. [11]). This is possible using state-of-the-art polishing methods. Nevertheless, the impact of the surface scattering strongly depends on the size of the resonator. Consequently, to measure $Q = 3 \times 10^{12}$ in a 100 μm diameter passive resonator, the surface roughness should be lower than 0.045 nm [11], which is smaller than values reported even for millimeter-size WGM resonators [20]. In our approach, for $\delta T_1 \to 0$, assuming an external coupling such as $\rho_1 \rho_2 = a$ and that the cold cavity Q factor is limited not by radiative or surface-scattering losses but by unsaturated absorption, we have $Q \approx \pi c T_1/(2\lambda_0)$, which shows that the size of the resonator does not have any detrimental effect on the Q-factor enhancement. Consequently, this principle could be extended to an on-chip integration of rare earth doped WGM microresonators [42] to obtain integrated microcavities with ultrahigh Q factors far beyond the state of the art, which could be used as a feedback loop in fully miniaturized ultrapure optical or microwave generators [5,7] for space applications where device size and volume are an issue. Note that, for our value of absorption $\alpha_0 \approx 0.1$ cm^{-1}, the unsaturated absorption limited Q factor is $Q_{\mathrm{mat}} = 6.2 \times 10^5$, which is orders of magnitude smaller than Q factors experimentally reported for on-chip WGM resonators [17]. The condition of cold cavity Q-factor limitation by material absorption would thus be easily obtained. Furthermore, this proof of principle shows that coherent or dispersive effects can be efficiently combined with photonic resonances in solid-state optical microresonators. Q-factor enhancement could thus also be observed

using EIT, CPT, or persistent spectral hole burning using WGM resonators doped with other ions [43]. These effects could be used instead of CPO to further increase the transparency of the dispersive medium. Finally, our experimental results can be interpreted not only as a slow-light enhanced cavity lifetime but also as cavity enhanced slow light. Indeed, the slow-light effect would not have been observable in a bulk sample made with the same erbium-doped glass. Because of the small optical depth, the phase shift accumulated during a single path would have been to be weak to be measured. Here, we take advantage of the high number of round-trips in the cavity ($\approx$700 for $\mathcal{F} = 1100$) to enhance the group delay. Extending this approach to CPO schemes involving three levels [44,45] or to other coherent mechanisms in erbium [46], it could be possible to obtain a fiber pigtail or arrays of miniaturized solid-state optical or quantum memories [47].

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