A Sub-$\lambda^3$-Volume Cantilever-based Fabry-Pérot Cavity

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We report on the realization of an open plane-concave Fabry-Pérot resonator with a mode volume of $\lambda^3/2$ at optical frequencies. We discuss some of the unconventional features of this new microcavity regime and show that the ultrasmall mode volume allows us to detect cavity resonance shifts induced by single nanoparticles even at quality factors as low as 120. Being based on low-reflectivity micromirrors fabricated on a silicon cantilever, our experimental arrangement provides broadband operation, tunability of the cavity resonance, lateral scanning and promise for optomechanical studies.

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Starting with the question of whether the excited-state lifetime of an atom can be modified, physicists have continuously explored ways to engineer the radiative properties of quantum emitters. Although the first experimental studies were performed in the near field of flat interfaces [1], the change of spontaneous emission is commonly associated with Purcell’s prediction that a cavity of quality factor $Q$ and mode volume $V$ can accelerate the radiation of a dipolar transition by $F_p = (3\lambda^3/4\pi^2)(Q/V)$ folds, where $\lambda$ is the wavelength in the corresponding medium [2]. Following this recipe, many clever resonator schemes such as high-$Q$ open Fabry-Pérot cavities (FPC), monolithic FPCs in form of pillars, whispering gallery mode resonators and photonic crystal cavities have been investigated for realizing large Purcell factors [3]. Nevertheless, routine and large modifications of the spontaneous emission process remain nontrivial because no cavity design has succeeded in providing the decisive ingredients of large $Q$, small $V$ (ideally down to its fundamental value of the order of $\lambda/2^3$), a facile way of tuning the cavity resonance, and compatibility with emitters of different materials. Of the various cavity geometries, open FPCs remain particularly attractive because they nicely lend themselves to the latter two criteria.

Efforts using open FPCs have usually pursued large $F_p$ via high $Q$s instead of low $V$. The downside of this approach is that the resulting narrow cavity linewidths do not allow simultaneous coupling to different transitions and are extremely sensitive to mechanically, thermally or stress-induced length changes. Furthermore, coupling to finite-sized particles in the condensed phase quickly spoils high $Q$s. In this Letter, we explore a new cavity design using a micromirror with a radius of curvature as small as 2.6 $\mu$m fabricated on a cantilever. The motivation for our work has several aspects. First, we want to realize a microcavity with an ultrasmall $V$ and low $Q$. The resulting $F_p$ leads to the modification of the spontaneous emission rate by more than one order of magnitude within a broad spectral bandwidth of 3 THz, which in turn allows the simultaneous coupling of various quantum transitions and differently detuned light fields to the cavity. The second novel feature of our arrangement is the compatibility of its high numerical aperture (NA) with the recent advances in efficient atom-photon coupling via tight focusing [4]. Third, the open character and scanning capability of our FPC provide the possibility of using it as a scanning cavity microscope [7, 8], which can be used for local field-enhanced spectroscopy [9] and sensing [10–12] of very different materials such as semiconductor quantum dots, carbon nanotubes, organic molecules, rare earth ions, and biomolecules. Fourth, the cantilever-based nature of our cavity makes it highly interesting for an unexplored regime of optomechanical investigations [13].

![Figure 1: (a) Schematics of the optical setup. OBJ: microscope objective; BS: beam splitter; HWP: half-wave plate; POL: linear polarizer; PD: photodiode; GNP: gold nanoparticle. The inset shows a zoom of the cavity region. (b) Scanning electron microscope image of four concave mirrors fabricated on a silicon pedestal at the end of a cantilever. (c) An atomic force microscope (AFM) cross section of a micromirror. (d) Numerical simulation of the intensity for the mode $q = 2$ of a cavity formed by a gold-coated micromirror and a flat distributed Bragg reflector marked by the dashed lines.](image-url)
The inset in Fig. 1(a) shows the schematics of our cavity made of a flat distributed Bragg reflector (DBR) and a micromirror fabricated by focused ion beam milling. Here, we started with an n-doped silicon cantilever that contained a pedestal (diameter 8 µm). The area of the micromirror to be milled was divided into pixels of diameter 5 nm. By controlling the ion dose for each pixel, we obtained a spherical surface profile, which was then polished in a final step. Figure 1(b) displays an electron microscope image of a cantilever pedestal with four mirrors of different radii of curvature, and Fig. 1(c) presents an exemplary topography cross section recorded with an AFM. A radius of curvature of R = 2.6 µm, root-mean-square surface roughness below 5 nm and an opening aperture diameter of 2.4 µm provide a concave mirror with numerical aperture NA = 0.68. Here, we follow the standard definition of NA = sin θ, where θ is the angle subtended by half the mirror aperture and its focal length given by R/2. We note that micromirrors have been previously reported using wet etching [14], laser ablation [15, 16] and focused ion beam milling [17], but the typical mirror radii of curvature remained well above ten micrometers, limiting both NA and V. To our knowledge, we provide the highest numerical aperture and smallest mode volume for a tunable microcavity reported to date.

The micromirrors were coated with 150 nm of gold followed by 50 nm of silicon dioxide as a protective layer, yielding a nominal reflectivity of 97%. The DBR consisted of 11 layers of TiO$_2$/SiO$_2$ stacks finished by a 22 nm layer of TiO$_2$ to place the field maximum at the mirror surface. The resulting structure had a total thickness of 2.14 µm with a band edge at 841 nm and reflectivity of 99.9% at λ = 785 nm. As displayed in Fig. 1(a), the microcavity was assembled with a piezoelectric transducer (PZT) stack for the axial displacement of the cantilever and a PZT scanner for the lateral positioning of the DBR.

Intuitively speaking, we want to operate the cavity close to the condition, where the rays from a strongly focused incident beam are retroreflected by the curved micromirror. It follows that a useful figure of merit for mode matching becomes the opening arc half-angle φ subtended by the mirror aperture (see the inset of Fig. 1(a)). We, thus, define $NA_{\text{eff}} = \sin \phi$ as an effective numerical aperture, which amounts to 0.4 in our experiment. We note that in practice the cavity has to be operated in the regime, where the cavity length $L < R$ in order to respect the stability diagram of a plane-concave FPC [18]. Figure 1(d) displays the intensity distribution obtained from numerical simulations of our experimental conditions [19].

To perform spectroscopy on the microcavity, we focused a laser beam at λ = 785 nm through a microscope objective with NA = 0.75, whereby the waist of the incident laser beam was adjusted to match $NA_{\text{eff}}$. We then scanned the axial position of the cantilever and monitored the reflected light on a photodiode. Figure 2(a) plots a typical cavity spectrum as a function of the change in L. Each mode is labeled with the cavity longitudinal mode number $q$, which usually determines the resonance wavelength λ according to $L = q\lambda/2$ for a simple FPC formed between two flat mirrors. For the fundamental transverse mode of a cavity with a curved mirror, however, one obtains [21]

$$L = \frac{\lambda}{2} \left[ q + \frac{1}{\pi} \arccos \left( \sqrt{1 - \frac{L}{R}} \right) \right].$$

Thus, when $R$ and $L$ become comparable one can observe notable deviations from the conventional FPC spectrum with a free spectral range of $c/2L$. Using Eq. 1, we determined $q$ and $L$ in a unique fashion from the cavity spectra recorded at two different wavelengths of 785 nm and 765 nm [19].

Figure 2: (a) Intensity of the light reflected from the cavity as a function of the cavity length change. Numbers below each resonance indicate the axial mode number. The small satellite resonances are attributed to higher order transverse modes. (b) Cavity Q (red) and finesse F (blue). Symbols: experiment; Curves: model. (c) Cavity volume V' (magenta) and Purcell factor $F_p$ (green). The lines serve merely as guides to the eye.

Figure 2(a) reveals that the full width at half-
maximum (FWHM) of the resonances of the different longitudinal modes \((\delta L)\) increase for higher \(qs\). To examine the performance of our FPC theoretically, we set up a model based on the propagation of a Gaussian beam between the two mirrors. We calculated the position and size of the beam waist \(w_0\) after each round trip, whereby we took into account the loss at the finite aperture of the curved mirror (about 0.1\% of the power per round trip for the \(q = 2\) mode) simply as a scalar factor \([19]\). The red curve in Fig. 2(b) shows the \(Q\) obtained from the cavity decay time and is in good agreement with the symbols, which represent the experimental values based on linewidth measurements. We do notice deviations for the highest mode orders, where \(L\) is large enough that our model is no longer valid and losses due to beam clipping become important. The nonmonotonous behavior of \(Q\) in Fig. 2(b) is the result of the competition between finite-aperture losses on the one hand and gain in the photon lifetime for larger \(L\) on the other.

The blue curve in Fig. 2(b) displays the finesse \(F\) computed as \(Q/q\), which again compares well with the experimentally measured values determined according to \((L_{q+1} - L_q)/\delta L\). Here too, the trend of \(F\) is at odds with the commonly encountered \(q\)-independent behavior for macroscopic cavities.

Another important parameter of the cavity is its mode volume \(V\). The magenta symbols in Fig. 2(c) plot \(V = \pi w_0^2 L/4\) of different modes deduced from the experimentally determined values of \(L\) and \(w_0\) \([19]\). For \(q = 2\) we find \(V \sim 0.5\lambda^3\), which corresponds to \(F_p \sim 17\) as shown by the green symbols in Fig. 2(c).

Having considered the basic features of our ultrasmall, broadband and tunable FPC, we now discuss experiments on coupling it to a point-like dipolar radiator. It is known that the introduction of a foreign object in the cavity adds to the overall optical path of the photons, leading to a red shift of the cavity resonance \([22, 23]\). In the case of a subwavelength nanoparticle, one arrives at the shift of the cavity resonance \(\Delta \nu\) given by

\[
\frac{\Delta \nu(r)}{\nu} = -\frac{R \epsilon(\alpha)}{2V} \frac{|E(r)|^2}{\max \| |E(r)|^2 \|}, \quad (2)
\]

where \(E(r)\) is the electric field at position \(r\) in the cavity \([22, 23]\). Here, \(\alpha\) is the complex electric polarizability of the particle, which is closely linked to its absorption and scattering cross sections \([24]\). In the quasi-static approximation

\[
\alpha = \frac{\pi D^3}{2} \left( \epsilon_p(\lambda) - \epsilon_m(\lambda) \right), \quad (3)
\]

where \(D\) is the particle diameter, and \(\epsilon_p(\lambda)\) and \(\epsilon_m(\lambda)\) are the dielectric functions of the particle and its surrounding medium, respectively.

One might have the intuitive expectation that the introduction of a foreign object into the cavity would cause losses, thus lowering its \(Q\). Although this is true in general, it has been shown that a nano-object can shift the cavity resonance without incurring a notable broadening \([23]\). Indeed, recently there has been a great deal of activity to exploit the frequency shift of a high-\(Q\) cavity for sensing nanoparticles such as viruses \([10, 25]\).

Assuming that the particle is placed at the field maximum of a cavity, Eq. (2) can be written as

\[
\frac{\Delta \nu(r)}{\nu} = -\frac{R \epsilon(\alpha) Q}{2V}, \quad (4)
\]

which expresses the ratio of a shift \(\Delta \nu\) to the linewidth \(\delta \nu\) of the cavity resonance profile. It is clear that a high \(Q\) facilitates the detection of a small frequency shift experienced by a narrow resonance line. In this work, we enhance the shift by using very small \(V\)s so that it can be detected even for broad cavity resonances.

![Figure 3](image_url)

Figure 3: (a) The shift of the \(q=2\) cavity resonance in THz as a function of the lateral position of a gold nanoparticle. (b) A cross section from (a) along the dotted line. (c) Cavity resonance shift measured for different longitudinal modes.

To investigate the effect of a nanoparticle on our cavity, we spin coated gold nanoparticles (GNP) of diameter 80 nm on the DBR with an inter-particle spacing larger than several micrometers. Figure 3(a) shows the cavity shift as a function of the lateral position of a GNP, and Fig. 3(b) displays a cross section from it. We observe a shift as large as 1 THz, equivalent to 30\% of the cavity linewidth, over a Gaussian lateral profile with FWHM \(\sim 770\) nm. We point out that the polarizability of the GNP at the wavelength of interrogation is equivalent to a virus particle with diameter of 200 nm immersed in water.

The red symbols in Fig. 3(c) plot the maxima of cavity resonance shifts for different longitudinal modes. As expected from Eq. (2), the effect of the particle rapidly
diminishes for higher $q$ modes and larger $V$. To verify the measured data quantitatively, we have fitted them with Eq. (2), leaving $\alpha$ as a free parameter. The blue curve shows the best fit obtained for $R\epsilon(\alpha) = 1.6 \times 10^6$ nm$^3$, which is 1.77 times larger than its nominal value for a GNP with a diameter of $80 \pm 6$ nm (as specified by the manufacturer), $\epsilon(\lambda)$ of gold obtained from Ref. [29], and $\epsilon_m = 1$. We point out that the exact knowledge of $\alpha$ for a given GNP is highly nontrivial. First, near-field coupling to the DBR surface modifies the GNP plasmon resonance and polarizability [27]. By using an analytical expression [28], we have verified that the polarizability of the GNP is expected to increase by 1.1 times in the presence of the DBR upper layer. Furthermore, variations in shape, the finite size of the particle and the resulting radiation damping effect [29] enter beyond the simple expression of Eq. (3).

Next, we turn to the effect of the nanoparticle on the cavity $F$ for various mode orders. Since the nanoparticle scatters some of the light out of the cavity mode and has a finite absorption cross section, one can expect a degradation of $F$. Figure 4(a) displays a lateral map of $F$ as a function of the GNP position in the $q = 2$ mode, and the top curve in Fig. 4(b) plots a cross section from it. We find that $F$ decreases by about 7% from 60 to 56 in the presence of the particle. However, as shown by the other plots in Fig. 4(b), this behavior is substantially changed for higher modes. In fact, we find that the particle can even improve $F$. For example, it is increased by about 10% from 6.5 to 7 for $q = 6$.

For the shortest cavities, the mirrors are so close that the light is efficiently captured after each round trip. In this case, absorption of the particle reduces the quality factor and finesse. As $L$ approaches $R$, the unperturbed resonator becomes less stable and $F$ is lowered (see Fig. 2(b)). In this regime, the addition of the GNP ameliorates the situation because it redirects some of the light that would be otherwise not captured by the finite NA$\text{eff}$ of the micromirror. In this sense, the GNP acts as a mode matching antenna that improves the coupling mirror reflectivity. To verify this hypothesis, we measured the amount of light that circulates inside the cavity by monitoring the power reflected from the DBR. Figure 4(c) plots the two-dimensional map of the coupling efficiency for $q = 2$, and Fig. 4(d) displays the cross sections for various modes. The similarity of the patterns of the two data columns in Fig. 4(b,d) shows that the GNP strongly influences the coupling of the incoming laser beam into the cavity mode. At this point, we note that one can also perform interferometric scattering measurements (iSCAT) to detect individual GNPs without the micromirror [30]. Using this method, we determined a FWHM of 690 nm for the focus spot of the incident beam in the absence of the cantilever [19]. This value agrees to within about 10% of the FWHM we found from Fig. 3(b), indicating that the cavity mode waist is well matched by the incoming spot size.

The cavity regime studied in this Letter has brought forth different phenomena that had not been encountered in previous works on microcavities. These include a strong dependence of the cold cavity $Q$ and $F$ on the longitudinal mode order $q$, a large numerical aperture and wavelength-sized mode waist, increase of cavity $F$ by a nanoparticle, the combination of a sub-$\lambda^3$ mode volume and frequency tunability, and the fact that one mirror sits on a cantilever. These features are very promising for a number of future studies ranging from biophysics to quantum optics.

In our current experiments, $(\lambda^3 Q/V)$ reaches a value of $\sim 250$. The green symbols in Fig. 2(c) show the Purcell factor for the different modes of our cavity. The maximum achieved value of $F_p = 17$ would be sufficient for turning a typical branching ratio of about 30% for the zero-phonon line of aromatic molecules to about 90%,
yielding a nearly perfect two-level system with important implications for efficient single-photon generation \[31\] and the realization of a single-molecule mirror \[3\]. Moreover, enhancement of the optical field in the cavity and of the radiation of a quantum emitter into its mode are very desirable for boosting nonlinear effects such as Raman scattering and four-wave mixing at the single-emitter level \[32\]. Here, a large cavity bandwidth is of crucial importance since simultaneous coupling to several wavelengths would be otherwise not possible. Furthermore, the low \( Q \) of the cavity and its resulting tolerance to mechanical instabilities, can be advantageous for use in atom \[33\] and nanoparticle trapping \[34\].

The cantilever-based nature of our experimental arrangement also holds great promise in the context of optomechanics. Let us consider a silicon cantilever of thickness 0.7 \( \mu m \), length 10 \( \mu m \) and width 5 \( \mu m \), yielding a mechanical oscillation frequency of about 5 MHz and mass of about \( 10^{-13} \) kg. A micromirror on such a cantilever forming a cavity length \( L = 1 \mu m \) would result in a nearly maximal field per photon \( \sqrt{\hbar \omega / 2 \epsilon_0 V} \) and an optomechanical coupling strength between the photon and a single phonon of \( g_{\text{opt}}^m/2\pi > 10^6 \) Hz, which is higher than the best reported values \[13, 35\]. Finally, we remark that although in this work we have pursued particularly low \( Q \) values, these can be readily increased by coating the curved mirror with highly reflective dielectric multilayers \[3\].

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