Whispering-gallery modes and light emission from a Si-nanocrystal-based single microdisk resonator

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Abstract: We report on visible light emission from Si-nanocrystal based optically active microdisk resonators. The room temperature photoluminescence (PL) from single microdisks shows the characteristic modal structure of whispering-gallery modes. The emission is both TE and TM-polarized in 300 nm thick microdisks, while thinner ones (135 nm) support only TE-like modes. Thinner disks have the advantage to filter out higher order radial mode families, allowing for measuring only the most intense first order modal structure. We reveal subnanometer linewidths and corresponding quality factors as high as 2800, limited by the spectral resolution of the experimental setup. Moreover, we observe a modification of mode linewidth by a factor 13 as a function of pump power. The origin of this effect is attributed to an excited carrier absorption loss mechanism.

OCIS codes: (230.1150) All-optical devices; (230.5750) Resonators; (140.3948) Microcavity devices; (250.5230) Photoluminescence.

References and links
1. L. Rayleigh, “Further applications of Bessel’s functions of high order to the whispering gallery and allied problems,” Philos. Mag. 27, 100 (1914).
2. K.J. Vahala, “Optical Microcavities,” Nature (London) 424, 839 (2003).
3. See, e.g. the review by V.S. Ilchenko and A.B. Matsko, “Optical Resonators With Whispering-Gallery Modes-Part I: Basics,” IEEE J. Sel. Top. Quantum Electron. 12, 76 (2006).
4. A.M. Armani, D.K. Armani, B.Min, K.J. Vahala, and S.M. Spillane, “Ultra-high-Q microcavity operation in H2O and D2O,” Appl. Phys. Lett. 87, 151118 (2005).
5. S.L. McCall, A.F.J. Levi, R.E. Slusher, S.J. Pearton, and R.A. Logan, “Whispering-gallery mode microdisk lasers,” Appl. Phys. Lett. 60, 289 (1992).
6. P. Michler et al., “Laser emission from quantum dots in microdisk structures,” Appl. Phys. Lett. 77, 184 (2000).
7. Zh. Zhang et al., “Visible submicron microdisk lasers,” Appl. Phys. Lett. 90, 111119 (2007).
8. K. Srinivasan, A. Stintz, S. Krishna, and O. Painter, “Photoluminescence measurements of quantum-dot-containing semiconductor microdisk resonators using optical fiber taper waveguides,” Phys. Rev. B 72, 205318 (2005).
9. L. Pavesi, L. Dal Negro, C. Mazzenoli, G. Franzo, and F. Priolo, “Optical gain in silicon nanocrystals,” Nature (London) 408, 440 (2000).
10. Towards the First Silicon Laser, NATO Science Series, edited by L. Pavesi, S. Gaponenko, and L. Dal Negro (Kluwer, Dordrecht, 2003).
11. A.S. Liu et al., “A high-speed silicon optical modulator based on a metal-oxide-semiconductor capacitor,” Nature (London) 427, 615 (2004).
12. H. Rong et al., “An all-silicon Raman laser,” Nature (London) 433, 292 (2005).
13. Q.F. Xu, B. Schmidt, S. Pradhan, and M. Lipson, “Micrometre-scale silicon electro-optic modulator,” Nature (London) 435, 325 (2005).
14. R.-J. Zhang, S.-Y. Seo, A.P. Milenin, M. Zacharias, and U. Gösele, “Visible range whispering-gallery mode in microdisk array based on size-controlled Si nanocrystals,” Appl. Phys. Lett. 88, 153120 (2006).
15. D.S. Gardner, M.L. Brongersma, “Microring and microdisk optical resonators using silicon nanocrystals and erbium prepared using silicon technology,” Opt. Mater. 27, 804 (2005).
16. L. Ferraioli, M. Wang, G. Pucker, D. Navarro-Urríos, N. Daldosso, C. Kompocholis, and L. Pavesi, “Photoluminescence of Silicon Nanocrystals in Silicon Oxide,” J. Nanomat. 2007, 43491 (2007).

1. Introduction

High quality monolithic resonators such as micro-disks, rings and toroids are triggering an intensive and rapidly evolving research. Such structures, in which the total internal reflection leads to circularly propagating optical modes, called whispering-gallery modes (WGM) [1], are extremely attractive both from device application and fundamental points of view [2]. Optically passive microdisks, based on transparent materials with negligible absorption losses, lead to extremely high quality factors (Q ~ 10^6 to 10^10), offering applications in spectroscopy and sensing [3, 4]. On the contrary, optically active resonator systems, such as III-V semiconductor quantum dot microdisk lasers, report active Q’s of 10^3–10^4 in the visible and near infrared wavelength range [5, 6, 7, 8].

The recent challenges in silicon photonics towards using nanocrystalline Si (nc-Si) as an integrated light source have boosted an intensive research in the last decade [9, 10, 11, 12, 13]. However, as an important cavity system, nc-Si-based microdisk structures have been little studied and only few works appear in the literature [14, 15]. In Ref.[14] quality factors of few hundreds have been reported for microdisk arrays of nc-Si/SiO_2 superlattices.

In this Letter we study the WGM emission properties of single microdisk resonators with an optically active disk material made of luminescent nc-Si embedded in SiO_x matrix. We report on subnanometer WGM resonances corresponding to quality factors as high as 2800 around the wavelength of 800 nm, which to our knowledge are the highest among the previously reported values in nc-Si-based systems. We demonstrate the importance of exciting a single resonator out of the mass-produced microdisk array in order to reveal the fine modal structure in the light emission. Additionally, we show an almost 13-fold narrowing of characteristic linewidths at lowest excitation power associated to an attenuation of excited carrier absorption losses.

2. Sample preparation and micro-PL setup

Our samples have been produced using standard silicon microfabrication technology. A Si rich silicon oxide (SRO) layer of 135 nm was deposited on top of crystalline silicon wafers from a mixture of silane (SiH_4, 65 sccm) and nitrous oxide (N_2O, 973 sccm) gases using a parallel-
plate plasma enhanced chemical vapor deposition (PECVD) chamber (deposition rate of $\sim 129$ nm/min). A successive one hour annealing in an $N_2$ atmosphere at $1100^\circ C$ leads to the formation of Si nanocrystals in the SiO$_x$ host (with $\sim 10\%$ of Si atoms in the nanocrystalline phase) [16]. Then the wafers were photolithographically patterned and dry etched anisotropically using CHF$_3$/SF$_6$ gas chemistry. This way arrays of microdisk structures with diameters ranging from 2$\mu$m to 10$\mu$m were formed. Isolation of the microdisks from the silicon substrate was realized by an isotropic wet etch of the latter, which formed mushroom-like devices.

Room temperature micro-PL measurements have been performed using the 488 nm line of an Argon laser (see Fig. 1). A long working distance objective was used to focus the laser beam on the samples. The microdisks close to the cleaved sample edge were excited vertically, while the WGM emission was monitored in the plane of disks, using collection optics (numerical aperture $NA = 0.13$). A polarizer was placed in the collection line to select transverse electrically (TE) or transverse magnetically (TM) polarized PL emission. Finally, the collected signal was sent to a spectrometer interfaced to a cooled silicon charge coupled device (CCD). The PL characterization was performed for microdisk arrays of various diameters, while for simplicity we focus our attention on the discussion of results obtained for the 8$\mu$m diameter resonators.

3. Whispering-gallery modes of single resonators

When hundreds of resonators are excited simultaneously through a large spot over the microdisk array, the observed emission lineshape is characterized by a convolution of the broad emission band ($\sim 300$ nm wide) of Si-nanocrystals with the WGM resonant spectrum of the microdisks (roughly 5 nm wide, $Q \sim 160$) [17]. Such low values of $Q$ can be attributed to an inhomogeneous broadening of peaks due to slight dispersion of microdisk diameters within the excited area [14, 18].

To demonstrate this, we focused the excitation onto an individual microdisk and recorded the resulting PL emission. In fact, from Fig. 2 one observes immediately the fine WGM structure of the single microdisk. The significant narrowing of the emission lines results in subnanometer full width at half maxima (FWHM), leading to quality factors of almost $3 \times 10^3$, limited by the spectral resolution of our micro-PL setup (higher precision measurements may reveal higher $Q$ values). We note that the trade-off between lower material absorption and weaker confinement of WGM modes at longer wavelengths on one side, and the limited emission band of
nanocrystals on the other, result in a limited bandwidth were high $Q$ factors can be observed.

To get some insight on the light confinement inside the microdisk resonator, we have performed effective index mode simulations of a slab waveguide structure with the same thickness ($d=135$ nm) and material refractive index (1.8 at 800 nm). For such choice of parameters, we have obtained a well confined TE mode ($n_{eff}=1.34$) while an almost-radiative TM mode ($n_{eff}=1.08$). Since these indices represent the upper limits for the $n_{eff}(\mathrm{TE, TM})$ of the 3D resonator structure, we therefore expect that our microdisk does not support a guided TM mode. This is confirmed both by experimental results and by 3D finite-difference time-domain simu-

Fig. 2. (color online) Measured TE-polarized WGM spectrum of an 8μm diameter microdisk is plotted together with the simulated peak positions for the first radial mode family (○). The predicted second mode family (⋆) is however absent in the measured spectrum because of being much less intense. (Inset) The bright spot in the photograph is the direct image of the visible PL emission of nc-Si from a single disk resonator.

Fig. 3. (color online) Typical TE-polarized WGM emission spectra from 8μm diameter (a) thin, $d=135$ nm and (b) thick, $d=300$ nm disk resonators. The corresponding insets show (a) the absence and (b) the presence of resonant whispering-gallery features for TM-polarization. Scanning electron microscopy images are shown respectively in panels (c) and (d). Note, that though the support pedestals have different top radii, in both cases they are small enough to not disturb the WGM characteristics.

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Fig. 4. (color online) The measured $Q$-factors at increasing pump power are plotted at three different wavelengths, reporting an order of magnitude variation between two extreme pump powers. The error bars reflect the accuracy limit of measured linewidths. The inset shows the WGM mode at $\lambda = 849$ nm at the lowest and at a high pump powers.

lations using a freely available software package [19]. Indeed, as one can see from the inset of Fig. 3(a), no characteristic WGM features can be observed for TM-polarized emission in the PL spectrum of a 135 nm-thick microdisk. For the TE polarization the simulations predict the existence of two radial families (radial mode numbers $p=1,2$). However, the measured TE-spectra (Fig. 2 and Fig. 3(a)) show only the $p=1$ modes, because the second family is much less intense. Thus, all the observed spectral peaks belong to the same family and their corresponding azimuthal mode numbers extend from $m=29$ (928 nm) to $m=42$ (710.5 nm), with an average mode spacing of $\sim 15$ nm.

We have prepared another series of microdisk arrays with a core thickness of $d=300$ nm in order to check the existence of TM-polarized WGM emission in thicker samples. In this case, slab calculations give $n_{\text{eff}}(\text{TM})=1.45$ (with $n_{\text{eff}}(\text{TE})=1.6$), suggesting a sufficient mode confinement for TM-polarized light. Figure 3(b) shows both TE- and TM-polarized (inset) PL spectra from an individual microdisk. As it was expected, now we clearly observe the fine features of WGM in the TM-polarization. On the other hand, higher order family modes with $p \geq 2$ can be resolved in the TE-spectrum.

4. Pump power induced mode broadening: The role of Excited Carrier Absorption

Finally, we address some issues related to the influence of pumping power on the WGM characteristics of our microdisks, in particular, the observed significant linewidth modification. Figure 4 reports the measured $Q$’s of few distinct resonances at $\lambda = 754$ nm, 768 nm and 849 nm ($m=39$, 38 and 33, respectively) of thin microdisks. We observe a monotonic ($\approx 13$-fold) decrease of $Q$ factors as the pump power increases from 1.25 to 100 mW. Such an impressive result requires a special attention.

The attenuating $Q$ factors suggest that at higher excitation powers we either introduce an additional loss source or enhance the existing ones. In a microdisk resonator, the total loss, resulting from different loss mechanisms, is expressed through the sum of inverse of possible limiting $Q$ factors:

$$Q^{-1} = Q_{\text{rad}}^{-1} + Q_{\text{mat}}^{-1} + Q_{\text{ssc}}^{-1} + Q_{\text{sa}}^{-1},$$

where the inverse of $Q_{\text{rad}}$, $Q_{\text{mat}}$, $Q_{\text{ssc}}$ and $Q_{\text{sa}}$ corresponds to radiation, material (bulk absorp-
nanocrystals will induce additional non-linear processes, such as the ECA itself (re-absorption of either a pump or emitted photon by an already formed exciton), Auger recombination and the saturation of the number of excitable photons to promote electrons to higher energetic levels in the nanocrystal conduction band. Such absorption events will enhance the cavity losses, causing the observed WGM broadening.

While under low pump conditions the ECA loss is expected to increase linearly with power, at high pump powers different phenomena can deviate this simple relationship; a number of processes, such as the ECA itself (re-absorption of either a pump or emitted photon by an already formed exciton), Auger recombination and the saturation of the number of excitable nanocrystals will induce additional non-linear $N(P)$ behavior. This non-linearity is clearly observable in all our experimental data (Fig. 4).

Assuming a power-dependent loss, $\alpha = \alpha_0 + \alpha^*(P)$, one can rewrite Eq. 1 as

$$Q^{-1}_\text{exp}(P) = \left( \sum Q^{-1}_\text{rad,ss,sa} + \frac{\lambda \alpha_0}{2\pi n_{\text{eff}}} \right) + \frac{\lambda \alpha^*(P)}{2\pi n_{\text{eff}}},$$

(2)

Here $\alpha_0$ stands for the material passive loss coefficient, which has been independently estimated from ellipsometric data and at $\lambda = 754$ nm is of the order of 30 cm$^{-1}$ or less [22]. The first two terms on the right part of Eq. 2 represent the inverse of $Q$ of the passive microdisk and, as mentioned above, can be considered as a constant term. With this, for the measured data $Q^{-1}_\text{exp}(P)$ a non-linear fitting function in the form $\alpha^*(P) \sim aP(1+bP)^{-1}$, based on the rate equation model [21], has been applied (Fig. 5).

The quality of the fit is further confirmed through the following procedure. We consider the fitting value for the passive $Q$ (the constant term in Eq. 2), the FDTD-simulated geometrical quality factor $Q_{\text{rad}} \approx 1.1 \times 10^4$ for 8 $\mu$m disks and negligible surface scattering/surface absorption losses, and back-calculate the material passive loss coefficient. Such obtained $\alpha_0 \approx 32$ cm$^{-1}$ is in very good agreement with the ellipsometric results.

The situation gets more complex when due to the cavity effect the spontaneous emission signal gets strong enough to affect the exciton population, as in the case of stimulated emission. When this becomes the dominant mechanism influencing the system loss, one expects that the absorption grows sublinearly with power (increasing transparency), leading to an inversion in the tendency of the $Q(P)$ curve (mode narrowing at high powers); hence, it would be possible to achieve net gain and eventual lasing at higher pump powers.

Thus, the possible presence of different loss mechanisms result in the observed complex behavior of $Q(P)$. In particular, we observe from Fig. 4, that all $Q(P)$ curves show a clear “shoulder” at increasing pump powers from 4 to 12 mW. A confident model and detailed studies for quantification of these phenomena are currently in course.

We stress that ECA will figure as the main limiting factor for a possible multiwavelength lasing from the nc-Si-based microdisk. The SRO material optimization (low-loss, positive material
gain) should play a key role for further enhancement of the observed $Q$-factors of few thousands. Even with low, while inhomogeneously broadened gain spectrum of nc-Si, microdisk resonators with similar $Q$‘s should be potential candidates to allow for a low-threshold laser action, in a similar way as in III-V semiconductor micro-disk/pillar devices [5, 6, 7, 23].

5. Conclusion

To conclude, we reported on PL emission properties of individual, optically active microdisk resonators with Si nanocrystals. We observed subnanometer whispering-gallery resonances in visible light emission with quality factors in excess of 2800 from single microdisk resonators. Moreover, the influence of pumping power on the WGM narrowing has been addressed, showing more than an order of magnitude enhancement of $Q$-factors due to an attenuation of pump-induced loss mechanisms. Both qualitative and quantitative analysis suggest that the excited carrier absorption stands behind the observed phenomenon.

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