Spectroscopy of 1.55 μm PbS quantum dots on Si photonic crystal cavities with a fiber taper waveguide

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We use an optical fiber taper waveguide to probe PbS quantum dots (QDs) dried on Si photonic
crystal cavities near 1.55 μm. We demonstrate that a low density (=100 μm−2) of QDs does not
significantly degrade cavity quality factors as high as ≈3 × 104. We also show that the tapered fiber
can be used to excite the QDs and collect the subsequent cavity-filtered photoluminescence, and
present measurements of reversible photodarkening and QD saturation. This method represents an
important step toward spectroscopy of single colloidal QDs in the telecommunications band.

The combination of low optical absorption and mature
device processing has resulted in the development of low
loss silicon photonic devices such as high quality factor (Q)
photonic crystal cavities (PCCs) operating in the technologi-
cally relevant 1.55 μm wavelength range.1–3 Silicon’s indi-
rect band gap represents a challenge in making light-emitting
devices and as a result there has been considerable interest in
developing hybrid systems integrating a light-emitting
material.4,5 Lead salt colloidal quantum dots (QDs) represent
one such approach. In addition, their atomic-like properties
suggest the potential for Si-based quantum information
processing in the single QD limit. In this work, we use col-
loidal PbS QDs as the active material to interact with Si
PCCs with resonances near 1.55 μm. Due to the long radia-
tive lifetime (≈700 ns (Refs. 7 and 8)) and small radiative
efficiency of these dried QDs (<1% (Refs. 7 and 9)), as well
as challenges associated with measuring low light levels with
InGaAs detectors,10 it is of the utmost importance to collect
as many emitted photons as possible. Previous studies of
PbS/PhSe QDs coupled to Si microcavities11–14 have relied
on free-space microphotoluminescence methods to pump and
collect the emission from moderately high-Q cavities (Q
≈103), and have generally operated at relatively high QD
densities, or else have sacrificed spectral resolution to
achieve the count rates needed to operate at a lower QD
density.15 In this work, we use an optical fiber taper waveguide2,16 to couple to the modes of high-Q PCCs (Q
≈104), thereby allowing for an efficient out-coupling mecha-
nism for PbS QD emission. We measure photoluminescence
(PL) from a low density (≤100 μm−2) of spun QDs and
show that the Q does not degrade due to QD absorption up
to Q = 3 × 104. We also measure photodarkening and saturation
of the QD emission into the cavity mode. This approach may
enable the future interrogation of cavity quantum elec-
drodynamics (cQED) in the PbS/Si system, in much the same way as has been demonstrated for epitaxial III-V QDs.17

The PbS QDs (Ref. 18) are chemically synthesized19 and
suspended in chloroform. As shown in the inset of Fig. 2(a),
the emission is centered near 1460 nm with a width of 100
nm due to a combination of size inhomogeneities and a large
homogeneous linewidth at room temperature. The solution is
further diluted with chloroform in a 1:200 mixture. Approximately 20 μL is spin-coated directly onto the substrate con-
taining PCCs, yielding an areal density of ≤100 μm−2 [inset
of Fig. 1(g)] as measured by a scanning electron microscope
(SEM). The PCCs measured [Figs. 1(a)–1(e)] are the well-
developed H1,20 L3,1 and multiheterostructure (MH) cavities,7 and have been fabricated in a 250 nm thick Si
device layer using standard silicon-on-insulator fabrication
methods. The devices are probed using an optical fiber taper
waveguide, which can be used to measure the spectral re-
sponse of the devices in transmission as well to collect PL.
Transmission measurements follow the approach of Ref.
2, where light from a swept wavelength external cavity diode

FIG. 1. (Color online) [(a)–(c)] SEM images of the H1, L3, and MH cavi-
ties, respectively. The lattice constants in (c) are {a1, a2, a3} = {410 nm, 415 nm, 420 nm}. [(d)–(f)] Transmission spectrum of the H1,
L3, and MH cavities before QD spin with fits (dashed). (d) and (f) were
taken with the taper in contact with the cavity, while (e) was taken with
the taper above the cavity. [(g)–(i)] Same as [(d)–(f)] but after QD spin. Inset
of (g) SEM image of QDs in a 256 × 173 nm2 area.
laser (1520 to 1630 nm) is sent through a variable optical attenuator and polarization controller before it is directed through the tapered optical fiber to an InGaAs photodiode. The taper and sample separation is controlled via x, y, and z stepper stages with 50 nm resolution, and the system is imaged under a 50× microscope objective. The measurement setup rests in a N₂-rich environment at room temperature to prevent irreversible photoxidation of the QDs (Ref. 21) and taper degradation.

This technique enables resonant spectroscopy of the cavity with and without the active material. In this way, we measured the cavity Q, before and after addition of the PbS QDs. Figures 1(d)–1(f) show a resonance of the H1, L3, and MH cavities in transmission without QDs, with measured Qs as high as 27 400. The extracted Q_{\text{eq}} values, which include intrinsic and parasitic losses, are 4900, 19 800, and 30 100, respectively. Figures 1(g)–1(i) show the cavity’s response in transmission with QDs with corresponding Q_{\text{eq}} = 4500, 23 200, and 29 500. For these low QD densities, the variation in the extracted Q_{\text{eq}} due to differences in taper position is greater than the loss induced by QD absorption, at least up to Q_{\text{eq}} \approx 3 \times 10^4. The ability to maintain high-Q in the presence of the QDs is promising for a number of potential applications, such as single QD cQED and low-threshold microcavity lasers.

For PL measurements, a 980 nm diode laser is coupled through a variable optical attenuator into the fiber taper, which is brought into contact with the devices. The transmitted signal is then directed through a long pass 1064 nm filter and into a grating spectrometer coupled with a liquid N₂ cooled InGaAs array. Spectra are recorded with a 180 s integration time under a typical excitation power of 100 μW. PL spectra from each cavity are shown in Fig. 2, including another mode in the MH cavity that did not appear in transmission [Fig. 2(c)]. The Q factors observed in PL are consistent with those seen in transmission measurements, though our spectral resolution is limited to ≈0.09 nm. We note that the cavity modes operate on the long wavelength tail-end of the QD distribution, as seen in the reference PL spectrum shown in the inset of Fig. 2(a) for an ensemble of QDs not in a cavity. This suggests the number of QDs interacting with the cavity modes may be significantly reduced with respect to the number that physically reside in the cavity, though a measurement of the QD homogeneous linewidth is needed to confirm this.

Using the transmission measurements in Fig. 1, we can estimate the efficiency η_o with which a cavity photon out-couples into the fiber taper. A QD’s out-coupling efficiency would then be the product of η_o with the fraction of QD radiation into the cavity mode. η_o is estimated from the on-resonance transmission level T_{\text{res}} as η_o = (1 − √T_{\text{res}})/2 where η_o represents collection in transmission. For the H1 cavity in Fig. 1(g), T_{\text{res}} = 0.381 so that η_o = 19.1%. A similar efficiency (T_{\text{res}} = 0.562 and η_o = 12.5%) has been measured when the taper is in contact with the L3 cavity [inset of Fig. 2(b)], while coupling to the MH cavity as shown in Fig. 1(i) yields a somewhat smaller value (T_{\text{res}} = 0.670 and η_o = 9.07%); fluctuations in the detected signal result in uncertainties in η_o of ±0.1%. These results generally compare favorably to calculated free-space collection efficiencies of ≈10% using high numerical aperture objectives, with the added advantage of direct collection into a single mode optical fiber.

Our experimental configuration also enabled measurement of photodarkening behavior previously observed in PbS QDs. In this case, PL from the MH cavity is directed through long pass filters at 1064 and 1400 nm and detected at an InGaAs single photon counting module (SPCM) (Ref. 10) with 2.5 ns gate width, 20% detection efficiency, and 5 μs dead time. As shown in Fig. 3(a), the PL is monitored continuously with a 0.6 s integration time while the 980 nm excitation source is turned on (P_{\text{drop}} = 154.0 μW ± 9.5 μW) and off. The PL clearly decays with time and requires an off time of at least 150 s to completely recover. This kind of photodarkening has been attributed to an average of single particle blinking where the overall ensemble PL decreases with time due to increasing numbers of emitters transitioning to a long-lived dark state. Figure 3(b) shows a normalized photodarkening trace taken under the same excitation conditions as in Fig. 3(a). The data has been fit with a stretched exponential,

\[ I(t) = I_{eq} \times (1 - \exp[-(t/T_o)]^{(1+1/r)} \times (1-\alpha/T_{\text{eq}}) \times \exp[-(t/T_{\text{eq}})]^{0.57 \pm 0.38}. \]

While the fit parameters T_o and α are consistent with literature, the actual physical parameters associated with QD blinking can only be determined with further...
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18Purchased from Evident Technologies and identified in this paper to foster understanding, without implying recommendation or endorsement by NIST.