Large Purcell enhancement of a quantum dot coupled to a circular grating in a charge tunable device

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The optical and spin properties of quantum dots position them as promising candidates for photonic quantum information processing and as building blocks of quantum networks. To efficiently utilize the quantum dot spin for such applications requires a good interface between the spin and arbitrarily polarized light, which can be provided by fabricated photonic structures with nearly degenerate polarization modes. Here, we study the optical properties of quantum dots in a charge tunable device (a p-i-n-i-n diode) coupled to such a promising photonic structure, the bullseye antenna. By leveraging the low charge noise associated with the device, we observe spontaneous emission lifetimes of such dots as short as 80 ps (a Purcell enhancement of ≈ 15). These emission rates are four times faster than the rates previously observed utilizing bullseye antennas with nearly degenerate polarization modes, and are close to the state-of-the-art emission rates observed on dots coupled to micropillars. In addition to Purcell enhancement, our fabricated bullseye antennas improve the optical interface with quantum dots by over an order of magnitude. Finally, due to the nearly degenerate polarization modes of the antenna, we are able to optically pump the spin of electrically charged quantum dots coupled to them. Coherently controlling the quantum dot spin while leveraging the improved optical interface offered by the bullseye antenna could upgrade the potential of such dots for quantum information processing.

In recent years, optically-active quantum dots have emerged as useful resources for photonic quantum technologies. Quantum dots emit single photons with high brightness and indistinguishability [1–8], which makes them promising as sources of single and entangled photons for photonic quantum computing [9–12]. In addition, these dots can be electrically charged with a single electron or a single hole, thereby offering a ground state spin qubit [13–19]. The coupling of such spins to single photons enables the generation of photon-photon interactions for quantum information processing [20], and could contribute to the ongoing efforts of establishing quantum networks [21–22].

To efficiently interface quantum dots with light often involves the coupling of the dots to fabricated photonic cavity structures [3, 5, 7, 23–27]. Such structures can increase the optical density of states in the environment of a quantum dot, thereby Purcell-enhancing the rate of spontaneous emission of single photons from the dot. In addition, photonic cavity structures that provide Gaussian far field emission patterns can improve the efficiency of exciting and collecting photons from the quantum dot via confocal optical setups.

A notable photonic cavity structure that efficiently interfaces quantum dots with light is a circular grating formed by etching a periodical pattern of rings from a sample that hosts single photon emitters, namely the bullseye antenna [7, 25, 29, 30]. To date, bullseye antennas have been used to optically interface single defects in diamond [29], as well as optically-active quantum dots [7, 25, 30]. In early implementations [25, 29, 30], the spontaneous emission lifetimes of the single photon emitters coupled to the antennas were no shorter than 350 ns. Recently, faster single photon emission rates were measured by introducing ellipticity to bullseye structures, which broke the degeneracy of their polarization modes [7]. However, this broken degeneracy reduces the potential of elliptical gratings toward applications that require the coherent control of the quantum dot spin, which require the excitation of the spin circularly polarized light [14–18, 19]. Furthermore, the impact of bullseye antennas on the optical properties of electrically charged dots at high magnetic fields, which offer such a spin qubit for quantum information processing [13–19], has yet to be explored.

Here, we fabricate bullseye antennas with nearly degenerate polarization modes to generate a large Purcell enhancement of quantum dots embedded in a charge tunable device, namely a p-i-n-i-n diode [31, 32]. By leveraging the low charge noise associated with the diode, we measure spontaneous emission lifetimes of quantum dots as short as ≈ 80 ps (a Purcell enhancement of ≈ 15), which are close to the state-of-the-art optical lifetimes measured on quantum dots coupled to micropillars [3, 4, 7]. In addition, the nearly degenerate polarization modes of the bullseye antennas allow us to optically excite and measure the Raman signal emitted from electrically charged quantum dots. Combined with the enhanced efficiencies of optically exciting the
quantum dot spin and collecting the emitted photons, the fabricated bullseye antennas offer a promising platform for quantum information processing utilizing electrically charged quantum dots.

We perform measurements on InAs quantum dots embedded in GaAs, deterministically charged by applying a DC bias voltage on a low charge noise p-i-n-i-n diode [Fig. 1(a)]. Under the application of an external magnetic field of \( B = 9 \, \text{T} \) perpendicular to the sample growth axis (Voigt geometry), we observe Zeeman splitting of a single optical transition of the dots into two and four transitions for uncharged and electrically charged quantum dots, respectively (see supplementary material). Our measurements of the emission lifetimes of quantum dots via these optical transitions exhibit decays on timescales of 1.1-1.3 ns (see supplementary material), consistent with previous observations on such dots [30].

To improve the optical interface of the quantum dots, we fabricate bullseye antennas by etching rings from the semiconductor membrane consisting of the p-i-n-i-n diode and a sacrificial AlGaAs-Si layer [white areas in Fig. 1 (a), see full fabrication details in the supplementary material]. Figure 1 (b) shows the SEM image of a typical fabricated structure, where the dark areas represent the etched material and the vertical and horizontal tapered lines represent "bridges" that prevent the structure from collapsing [29]. A finite-difference-time-domain simulation of the far field emission of light from the antenna results in a Gaussian pattern with waist smaller than the numerical aperture (0.68) of collecting photons via the objective lens of our confocal optical setup. Such matching between the far field emission pattern and the numerical aperture highlights the potential of the antenna in efficiently exciting and collecting photons from quantum dots coupled to it. In addition, the simulated far field emission pattern is polarization independent. This polarization independence positions bullseye antennas promising for the coherent control of the spin of electrically charged quantum dots coupled to the antenna, which typically requires pulses of circularly polarized light [13, 18, 19].

To study the impact of the bullseye antennas on the optical properties of quantum dots, we locate such an antenna coupled to two separate uncharged dots [Fig. 2 (a)]. By applying laser light with energies above the band gap of InAs (i.e., an above-band excitation), we generate charge carriers in the wetting layer of the sample that induce spontaneous photon emission from the antenna [solid blue line in Fig. 2 (a)]. The measured quality factor of \( \approx 1070 \) of the antenna agrees with our theoretical predictions from finite-difference-time-domain simulations. After the observation of photon emission from the antenna, we tune the voltage of the p-i-n-i-n diode to the plateau of two quantum dots (i.e., this voltage supports the dots while minimizing environmental charge noise). We identify the spontaneous emission of photons from the dots [dashed red lines in Fig. 2 (a)] by reducing the power of the above-band laser to eliminate the photoluminescence from the antenna. Sweeping the external magnetic field from \( B = 0 \) to \( B = 9 \, \text{T} \) (supplementary material) verifies that these dots are two separate and uncharged dots, with one dot (labeled "Dot 1") on spectral resonance with the central frequency of the antenna, and the other (labeled "Dot 2") slightly detuned from this frequency.

Figure 2 (b) compares the spontaneous emission lifetimes of the two dots in the bullseye antenna to a similar representative measurement on a quantum dot in the bulk. In the presence of the antenna, the emission lifetime shortens from \( \approx 1.2 \, \text{ns} \) [dotted green line in Fig. 2 (b)] for bulk dots down to \( \approx 160 \) ps (a Purcell enhancement of \( \approx 7.5 \)) for the dot spectrally detuned from the antenna, and down to \( \approx 80 \) ps (a Purcell enhancement of \( \approx 15 \)) for the dot resonant with the antenna. The latter lifetime is close to the state-of-the-art lifetimes measured on quantum dots embedded in micropillars [9, 4, 7], and are shorter by a factor of \( \approx 4 \) than those previously measured on quantum dots in bullseye antennas with nearly degenerate polarization modes [23, 30]. We attribute this improvement over
FIG. 2. (Color online) (a) Photoluminescence spectra of a bullseye antenna (solid blue line) and two uncharged quantum dots coupled to the antenna (dashed red line). (b) Time-resolved measurements of the optical lifetimes of quantum dots in bulk (dotted green line), as well as of two quantum dots coupled to a bullseye antenna labeled in (a) as ”Dot 1” (solid blue line) and ”Dot 2” (dashed red line).

previous work to two main factors. First, our fabrication of bullseye structures involves the etching of a sacrificial layer below the antenna [round cornered white rectangle in Fig. 1 (a)], whereas the rings of the bullseye structures in previous fabrications [25] [30] were not etched all the way down to this layer. While such partial etching improves the collection efficiency of photons scattered from the sample, it reduces the quality factor of the bullseye antenna [25], thereby providing a smaller Purcell enhancement compared to the ones observed by us after etching the sacrificial layer. The second factor that contributes to the enhanced optical emission rate is the deterministic charge capabilities provided by the p-i-n-i-n diode, which result in a reduction of the charge noise in the antenna’s environment [31] [32]. This low charge noise reduces the effects of spectral wandering of the antenna that may degrade the Purcell enhancement of the emission of the quantum dots coupled to it.

We next study the impact of the fabricated bullseye antennas on the optical properties of quantum dots charged with single electrons, which offer a useful spin qubit for quantum information processing. Using a series of ultrashort above-band laser pulses (i.e., they are much shorter than the optical emission rates of the dot), we locate the optical transitions of such a dot coupled to another bullseye antenna [Fig. 3 (a)]. Sweeping the external magnetic field (see supplementary material) shows that these four transitions at $B = 9$ T emerge from the Zeeman splitting of a single optical transition at $B = 0$, indicating their origin from a charged dot. At $B = 9$ T, the $\sim 1$ nm spectral detuning of these optical transitions from the main resonance of the antenna allows us to clearly distinguish between the emission of photons from the dot with the emission from the antenna. Compared to dots in the bulk, the laser power required for the saturation of the photoluminescence signal from the quantum dot in the antenna is an order of magnitude weaker, and the intensity of this signal is $\approx 25$ stronger. These outcomes indicate that the antenna provides an efficient optical interface for exciting and collecting light from the dot. Furthermore, as photons emitted via different optical transitions of charged quantum dots have polarizations orthogonal to each other, our observation of photoluminescence from all four transitions indicates the ability to excite the dot with arbitrarily polarized light.

We note, however, that the photoluminescence signals collected from the horizontally polarized transitions are weaker than the ones collected from the vertically polarized transitions. These differences in the intensity arise due to a small spectral splitting of polarization modes of this particular antenna due to fabrication imperfections (see supplementary material). Despite such quantitative differences, the ability to access quantum dots with light beams orthogonal to each other is crucial for the realization of pulse sequences for coherently controlling the quantum dots spin for quantum information processing, which typically require circularly polarized light [13] [18] [19].

To further emphasize the potential of controlling the quantum dot spin in the bullseye antenna, we use laser pulses resonant with one of the optical transitions of the dot to optically pump the spin. Varying the free evolution time between these pulses and measuring the emission of Raman signal from the dot results in the saturation behavior depicted in Fig. 3 (b). The sharp peak of the Raman signal emitted under the application of the first pulse indicates the optical initialization of the spin to one of its ground states. After such an initialization of the spin, the application of additional pulses that pump the same optical transition should not induce any Raman signal. Experimentally, however, the pulses induce undesired Raman signals that saturate for the free evolution time of $\approx 30$ ns between the
FIG. 3. (Color online) (a) The photoluminescence spectrum of an electrically charged quantum dot coupled to a bullseye antenna, under an ultrafast pulsed above-band excitations with an average optical power of 10 $\mu$W. The number of photons collected from the quantum dot is $\sim 25$ times larger than the number of photons collected from dots in the bulk under 100 $\mu$W above-band excitations. (b) Raman signals collected from the quantum dot under resonant excitation pulses at varying times. The sharp peaks indicate the optical pumping of the spin, and the increasing heights of these peaks with time indicate the relaxation of the quantum dot spin on a timescale of $\sim 30$ ns.

This saturated Raman signal represents the relaxation of the quantum dot spin, which reduces the spin initialization fidelity. This spin relaxation is dominated by two physical mechanisms. First, the natural spin relaxation time of quantum dots in our sample is limited to several tens of ns due to the tunneling of the electron confined in the dot. These natural relaxation times can be further extended by orders of magnitude by modifying the tunnel barriers (GaAs layers) of the diode [33, 34]. The second cause for the short spin relaxation time observed here is the spectral proximity of the resonant wavelength of the bullseye antenna ($\sim 905$ nm) to the InAs band gap. Given this spectral proximity, pumping an optical transition of the dot coupled to the antenna may lead to a residual above-band pumping of both spin states, thereby reducing the spin population. This residual pumping can be mitigated by designing and utilizing bullseye antennas with larger dimensions. The mitigation of both natural and laser-induced spin relaxation mechanisms could enable high fidelity spin control of the quantum dot spin using low laser powers, thereby upgrading the potential of these quantum dots for quantum information processing.

To conclude, we fabricate bullseye antennas to improve the optical properties of InAs quantum dots in a charge tunable device. By leveraging the low charge noise associated with the device, the dots exhibit single photon emission lifetimes as short as 80 ps. The antennas also increase the efficiency of exciting and collecting photons from the dots by over an order of magnitude, and support the optical pumping of the quantum dot spin. While the optical lifetimes measured here could be further shortened by utilizing resonant pulse trains [5], the optical interface could be improved by utilizing a distributed bragg reflector, and the fidelity of pumping the quantum dot spin can be improved by modifying the physical dimensions of the antenna and the diode. Combined with the nearly degenerate polarization modes provided by the bullseye antenna, such improved optical interfaces could enable the coherent manipulation of quantum dot spin for quantum information processing and quantum sensing.

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