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Coherent Spin-Photon Interface with Waveguide Induced Cycling Transitions

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Solid-state quantum dots are promising candidates for efficient light-matter interfaces connecting internal spin degrees of freedom to the states of emitted photons. However, selection rules prevent the combination of efficient spin control and optical cyclicity in this platform. By utilizing a photonic crystal waveguide we here experimentally demonstrate optical cyclicity up to $\approx 15$ through photonic state engineering while achieving high fidelity spin initialization and coherent optical spin control. These capabilities pave the way towards scalable multiphoton entanglement generation and on-chip spin-photon gates.

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Single solid-state spins play an important role in modern quantum information technologies [1–3]. A key resource is a coherent light-matter interface connecting spins and photons compatible with high fidelity spin manipulation and long distance distribution of quantum states [4,5]. Self-assembled semiconductor quantum dots (QDs) are currently considered one of the most promising systems thanks to their high photon generation rate, good optical and spin coherence properties, and efficient integration into photonic nanostructures [6]. The latter is important for realizing strong light-matter interaction and high collection efficiencies [7,8].

An application particularly well suited to QDs is the deterministic generation of multiphoton entangled states such as photonic cluster states [9]. These maximally entangled states have applications in measurement-based quantum computing [10] and quantum repeaters [11,12]. So far, one-dimensional cluster states generated with QDs have shown genuine 3 qubit entanglement with an extrapolated 5 photon entanglement [13]. A recent protocol based on time-bin encoded photonic qubits has been put forward allowing scaling up to tens of high-fidelity entangled photons for experimentally relevant parameters [14,15]. Crucially, this protocol requires an optical transition with high cyclicity, i.e., a transition where the excited state decays selectively to one of the two ground states, thereby preserving the spin.

In the context of QDs, the cyclicity corresponds to the ratio between the decay rates of the vertical transitions and the diagonal transitions, see Fig. 1(a). High cyclicity can be achieved by operating the QD in an out-of-plane magnetic field (Faraday geometry) where the diagonal transitions are only weakly allowed [16]. This configuration, however, obstructs fast all-optical spin control. For this reason, an in-plane magnetic field (Voigt geometry) has been indispensable for achieving spin control as demonstrated in bulk media [17–19], cavities [20–22], and nanobeam waveguides [23]. In this geometry, vertical and diagonal transitions possess equal magnitude, thus precluding cycling.

FIG. 1. (a),(b) Energy level diagram of a (a) negatively charged and (b) positively charged QD in an in-plane magnetic field. $\uparrow$, $\downarrow$ indicate electron spins and $\uparrow$, $\downarrow$ indicate hole spins. The four linear dipoles have equal decay rates in bulk but are selectively enhanced by the waveguide. (c) Scanning electron microscope image of the two-sided waveguide containing a PCW section, QD, and grating couplers. Fluorescence is always collected from the left coupler. Circular inset shows the ideal orientation of the linear dipoles for selective waveguide coupling.
transitions. However, cyclicity may be induced by a nanostructure through selective enhancement of the optical transitions. Such an enhancement has recently been demonstrated in cavity systems including single rare-earth ion spins [24]. QDs coupled to photonic crystal cavities [20,21,25] and micropillar cavities [14,22] also exhibit selective enhancement, but require tuning the QD into cavity resonance.

Here, we report the first realization of QD cycling transitions in the Voigt geometry due to the selective enhancement provided by a photonic crystal waveguide (PCW). The mechanism is fundamentally broadband as it relies on the orthogonally polarized dipoles having vastly different projections along the local electric field, as quantified by the projected local density of optical states in the PCW [6]. This approach has several advantages as it allows high cooperativity of multiple nondegenerate optical transitions, does not require substantial energy splittings of the optical transitions, and allows direct integration into photonic circuits. We measure a cyclicity of 11.6 (14.7) for the negative (positive) charge states of the same QD despite a 3.8 nm spectral separation. In addition, we achieve 98.6% spin initialization fidelity, $T_2 = (21.4 \pm 0.7)$ ns spin dephasing time, and all-optical spin control on the positively charged QD. These achievements together with the efficient photon collection of the PCW make our platform highly advantageous for the generation of multiphoton entangled states and the implementation of deterministic spin-photon quantum gate operations [26,27].

The nanostructure under consideration is a PCW made from a suspended GaAs membrane and connected to grating couplers [28] at either ends, see Fig. 1(c). The membrane comprises a $p$-$i$-$n$ diode grown in the $z$ direction with the intrinsic layer containing self assembled InAs QDs [29]. Applying a forward bias voltage deterministically charges the QD with a single electron (XM configuration), whereas additional optical induction allows the creation of a metastable hole state (XP configuration) with $> 16 \mu$s lifetime [30].

Applying an in-plane magnetic field results in a four level spin system for the XM [Fig. 1(a)] and XP [Fig. 1(b)]. The XM (XP) systems comprise two Zeeman split ground states with a single electron (hole) spin and two Zeeman split trion states containing a magnetically active hole (electron) and a singlet of two electrons (holes). The selection rules, which are functionally identical for XM and XP, result in two A systems, each containing an $x$- and a $y$-polarized linear dipole. While the associated radiative decay rates $\gamma_x$ and $\gamma_y$ are identical in bulk [36] the photonic environment of the PCW may selectively enhance and suppress the orthogonally polarized transitions, yielding a cyclicity $C = \gamma_y/\gamma_x \geq 1$. The decay rates can be further decomposed into a waveguide (wg) and radiative (rad) component, $\gamma = \gamma_{\text{wg}} + \gamma_{\text{rad}}$, $\gamma = \{x, y\}$. $\gamma_{x,\text{rad}}$ and $\gamma_{y,\text{rad}}$ are determined by the coupling to a continuum, are similar in magnitude, and generally strongly suppressed throughout the PCW [37]. In contrast, $\gamma_{x,\text{wg}}$ and $\gamma_{y,\text{wg}}$ are determined by the projected coupling onto the single polarized waveguide mode and can thus vary between zero and a highly enhanced rate given by the high optical density of states. This is the origin of the high cyclicity in the PCW which can be sensitively controlled by the position of the QD [37].

We now determine the cyclicity of the XM and XP systems with the two-color pump and probe pulse sequence in Fig. 2(a). For XM, a narrow band laser pulse with fixed power prepares $|\uparrow\uparrow\rangle$ by driving $|\downarrow\rangle \rightarrow |\downarrow\downarrow\uparrow\rangle$. The probe pulse performs the opposite operation, driving $|\uparrow\rangle \rightarrow |\uparrow\downarrow\uparrow\rangle$ and preparing $|\downarrow\rangle$. Both pulses are $y$ polarized to...
minimize coupling to the $x$ transitions. In the limit of $C \gg 1$ the rate of optical spin pumping $\gamma_{\text{osp}}$ saturates according to

$$\gamma_{\text{osp}} = \gamma_x \int_{-\infty}^{\infty} \frac{\Omega_p^2}{2\Omega_p^2 + \gamma_0^2 + 4\Delta_n^2} N(\Delta_n; \sigma) d\Delta_n,$$

where $\gamma_0 = \gamma_x + \gamma_y$ is the excited state decay rate, $\Omega_p = \gamma_0 \sqrt{P/P_{\text{sat}}}$ is the probe Rabi frequency, $P$ is the optical power, and $P_{\text{sat}}$ is the saturation power. For completeness, we include slow spectral diffusion via the detuning $\Delta_n$, which is drawn from a normal distribution $N(\Delta_n; \sigma)$ with standard deviation $\sigma$, although the effect on the cyclicity estimate turns out to be minor (< 3%). By varying the probe power and fitting the fluorescence histograms [Fig. 2(b)] a set of spin pumping rates are obtained. These rates are plotted in Fig. 2(c) and fitted with Eq. (1). A characterization (Supplemental Material [30]) of the XM yields $\gamma_0^{(\text{XM})} = (3.07 \pm 0.06)$ ns$^{-1}$ and $\sigma^{(\text{XM})}/(2\pi) = 140$ MHz. The pumping measurement then yields $\gamma_0^{(\text{XM})} = (0.243 \pm 0.005)$ ns$^{-1}$ and a cyclicity of $C^{(\text{XM})} = (\gamma_0^{(\text{XM})} - \gamma_x^{(\text{XM})}) / \gamma_x^{(\text{XM})} = 11.6 \pm 0.4$. The XP cyclicity is measured with the same method but with the inclusion of a hole initialization pulse [38]; see Fig. 2(a). Here we find $\sigma^{(\text{XP})}/(2\pi) = 345$ MHz, $\gamma_0^{(\text{XP})} = (2.48 \pm 0.02)$ ns$^{-1}$, $\gamma_x^{(\text{XP})} = (0.158 \pm 0.002)$ ns$^{-1}$, and a cyclicity of $C^{(\text{XP})} = 14.7 \pm 0.2$. Hence, XM and XP both demonstrate substantially increased cyclicity owing to both an inhibited $\gamma_x$ and an enhanced $\gamma_y$ compared to the expected bulk values [36]. This enhancement occurs for both XM and XP despite a spectral separation of 3.8 nm, demonstrating the broadband enhancement provided by the waveguide.

High-fidelity spin initialization is the essential starting point for applications of spin-photon interfaces. By analyzing the steady state fluorescence at the end of the spin pumping histograms we estimate lower bounds of the spin initialization fidelities $F_x^{(\text{XM})} = \langle \downarrow | \rho | \downarrow \rangle = 99.1\%$ and $F_y^{(\text{XM})} = \langle \uparrow | \rho | \uparrow \rangle = 98.6\%$, which are the highest values so far reported in photonic nanostructures [14,20,21]. In contrast to cross polarization experiments, our laser polarization control allows us to avoid driving the $x$-transitions which otherwise reduce the initialization fidelity through re-pumping. The $1/e$ spin pumping time when driving a $y$ transition is limited to $2/\gamma_y^{(\text{XP})} = 12.7$ ns, although driving an $x$ transition would result in significantly faster initialization at the cost of reduced fidelity owing to the $x$ transitions’ increased frequency overlap.

To further investigate the origin of the cyclicity we explicitly probe the coupling between the XM dipoles and the mode of the PCW. First, we analyze the spontaneous emission spectrum following continuous wave $p$-shell excitation. This method allows effective elimination of laser background (see Methods) and population of both negative trions. The emission is analyzed using a scanning Fabry-Perot cavity, revealing all four optical transitions, see Fig. 3(a). By fitting the spectrum we extract the intensity ratios $I_{y1}/I_{x1} = 21.8 \pm 0.5$ and $I_{y2}/I_{x2} = 20.3 \pm 0.6$, which do not differ significantly (1.9$\sigma$ deviation) and quantify the trions’ strong preference to decay into the PCW via the $y$ transitions. Taking the average, we estimate a waveguide-coupling asymmetry of $A^{(\text{XM})} = \gamma_{\text{wg}}^{(\text{XM})} / \gamma_{\text{wg}}^{(\text{XP})} = 21.1 \pm 0.4$. It should be stressed that this measurement only probes the dipole to waveguide coupling whereas the cyclicity also receives contributions from radiative modes causing it to be lower.

Next, we probe the coherent interaction between the four QD dipole transitions and the waveguide mode by measuring waveguide transmission. Previous experiments on charged QDs in waveguides were conducted on XM in the Faraday configuration where single photon switching was demonstrated [39]. In the present study we observe all four XM dipoles in the Voigt geometry. To avoid optical spin pumping we operate in the cotunneling regime where tunneling to the back contact of the diode provides a randomization of the electron spin with the rate
\(\kappa_{0} / (2\pi) \approx 1-10 \text{ MHz} \ [16]. \) In the limit \( \kappa_{0} \gg \gamma_{\text{osp}} \) spin flips randomize the spin in a thermal state \( \hat{\rho} \approx 0.53 \langle \uparrow \rangle \langle \uparrow \rangle + 0.47 \langle \downarrow \rangle \langle \downarrow \rangle \) given the temperature \( T = 4 \text{ K} \) and a 2 T magnetic field. Figure 3(b) shows the XM transmission when operating in the cotunneling regime. We observe a 0.66 GHz linewidth (1.35 \times \text{ natural linewidth}) and transmission dip amplitudes up to 39% where a factor \( \approx 2 \) reduction in amplitude stems from the thermal mixture. This pronounced transmission spectrum directly demonstrates the coherent nature of the spin-photon interface, which is required for applications such as single photon transistors [40–42], deterministic Bell state analyzers [27], and quantum gates [43,44]. Fitting the spectrum with an empirical model [30] yields a pronounced \( \approx 27 \) ratio between the \( y \) and \( x \) transmission dips owing to the selective waveguide coupling. Strikingly, rotating the magnetic field does not alter the dip amplitudes, suggesting an insensitivity of the dipole orientations to the magnetic field. This has previously been observed in Stranksi-Krastanov grown QDs and is attributed to an anisotropic QD shape or strain profile [45]. However, based on our previous observation of near equal \( x \) and \( y \) dipole oscillator strengths in bulk experiments [46] and on a moderate fine-structure splitting of the neutral exciton [30], no significant oscillator strength asymmetry is expected.

It is of course instructive to consider the limiting factors of the cyclicity. In a related paper [15] we predict that the cyclicity strongly depends on the placement of the QD within the PCW. Additionally, by approaching the PCW band edge one increases the group index, thereby enhancing \( \gamma_{\text{y,wp}} \) and in extension \( C \). Simulations [47] predict \( C = 144 \) being achievable at a group index of 56, corresponding to a realistic Purcell enhancement of about 10 [48]. We attribute our observed cyclcities to a nonoptimal position of the QD, a slightly lower group index, and possibly an undesirably large free space coupling as suggested by \( A^{\text{XM}} > C^{\text{XM}} \). Consistently realizing high cyclicity thus requires correct spatial positioning of the QD, correct orientation between the QD dipoles and the PCW mode and engineering of the PCW band edge. Deterministic fabrication of a PCW relative to a QD has already been demonstrated [49] so additional control over the PCW rotation is feasible.

Finally, to properly evaluate our system’s usefulness for quantum information processing applications we consider quantum control of the hole spin due to its increased coherence time over the electron spin [50,51]. We employ the Raman spin control scheme recently demonstrated in Ref. [19] which allows flexible phase control of the spin and elaborate electronically defined pulse sequences. A circularly polarized CW laser is red detuned by \( \Delta_{R} \) from the main optical transitions and amplitude modulated at frequency \( \Delta_{D} / 2 \) resulting in two sidebands matching the ground state splitting. The optical field then creates an effective coupling between \( \langle \uparrow \rangle \) and \( \langle \downarrow \rangle \) with Rabi frequency \( \Omega_{\text{MW}} \propto P_{R} / \Delta_{R} \), where \( P_{R} \) is the Raman laser power, see Fig. 4(a). By varying the duration of the pulse we observe clear Rabi oscillations between \( \langle \uparrow \rangle \) and \( \langle \downarrow \rangle \), see Fig. 4(b). We follow the model in Ref. [19] to estimate a \( F_{\pi} = 0.91 \) \( \pi \)-pulse fidelity upper bound. Applying the Ramsey pulse sequence in Fig. 4(c) yields the free induction decay of the hole spin, which is modeled according to Ref. [19]

\[
I(T) = I_{0} e^{-\left(\pi / T_{2}\right)^{2}},
\]

and yields a dephasing time of \( T_{2} = (21.4 \pm 0.7) \text{ ns} \). This is long compared to the emitter lifetime, \( T_{2} \gamma_{0}^{\text{XP}} = 54 \), thus allowing a considerable number of photons to be emitted within the spin dephasing time. We note that this \( T_{2} \) is on par with the performance found only in bulklike samples and at higher magnetic fields [38,52], demonstrating the capability of overcoming deteriorating noise processes in the nanostructures despite the nearby proximity of the QD.
to surfaces. For comparison, previous experiments in nanostructures reported a $T_2^* \approx 2.11$ ns of a hole spin in micropillar cavities [14] and below one nanosecond for electron spins in planar photonic cavities [20,21].

To summarize, we have successfully demonstrated how the photonic environment of a QD can be engineered to provide broadband selective enhancement of optical transitions resulting in cycling transitions in the Voigt geometry. The PCW accomplishes this without the need for high magnetic fields and QD tunability otherwise required by a cavity. The ability to excite the QD via a side channel magnetic fields and QD tunability otherwise required by a cavity. The ability to excite the QD via a side channel

The demonstrated $C_{XPP} \approx 15$ enables generation of multiphoton cluster states using time-bin encoding [15,53] which is insensitive to the spin dephasing $T_2^*$. Given the already demonstrated $\eta = 7\%$ collection efficiency [8], an increased cyclicity would serve to increase the single shot readout fidelity [54] from 53% ($C = 1$) to 75% ($C = 14.7$) and possibly as high as 95.5% ($C = 144$). Finally, the demonstration of a coherent spin-photon interface will be of immediate use for spin-photon and photon-photon quantum gates mediated by the efficient coupling between the QD and the PCW.

The data presented in this manuscript are available at [55].

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[30] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.126.013602 for hole lifetime measurements and modeling based on Ref. [31], for details on resonant transmission modelling which include Ref. [32], and for spectroscopy of the neutral exciton finestructure splitting which is compared against Refs. [33–35].