Polarization correlated photons from a positively charged quantum dot

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Polarized cross-correlation spectroscopy on a quantum dot charged with a single hole shows the sequential emission of photons with common circular polarization. This effect is visible without magnetic field, but becomes more pronounced as the field along the quantization axis is increased. We interpret the data in terms of electron dephasing in the $X^+$ state caused by the Overhauser field of nuclei in the dot. We predict the correlation timescale can be increased by accelerating the emission rate with cavity-QED.

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Spins in quantum dots (QDs) provide a promising platform for manipulating and storing quantum information in the solid state. Optical measurements have demonstrated spin preparation [1, 2] coherent spin control [3] and electron-spin – photon entanglement [4, 5]. There are also proposals for achieving photon entanglement [6] and non-destructive measurement of photons [7] using charged QDs. However, the time evolution of the carrier spin is unavoidably affected by the $10^4 - 10^5$ nuclei in the dot, all with non-zero spin. One example of the utility of the electron-nuclear interaction is its use in spin pumping the hole into the spin down state in zero external field, by pumping on the spin up state of $X^+$ [2]. From a fundamental point of view then, the hyperfine interaction provides an interesting system for manipulating a mesoscopic nuclear ensemble and observing its dynamics.

It has now been established that the electron-nuclear hyperfine interaction is dominated by the contact interaction and is isotropic in a QD [8]. The dynamics of this interaction manifests itself in studies of polarized photo-luminescence [9, 10]. Contrastingly, the hole’s $p$-like wave-function has a node at each nucleus leaving the dipole-dipole interaction between the hole and nuclear spins to dominate [11]. This interaction has a strength one order of magnitude below that of the electron [12, 13]. Thus, there has been interest in using the hole-spin as quantum bit with reduced decoherence. Direct measurements of the hole spin relaxation time in a vertical magnetic field, $T_1^h$, have shown it is hundreds of microseconds [2, 14]. Without applied magnetic field some experiments suggest the hole spin $T_1^h$ time is 13 ns [8]. Several studies have now estimated the hole dephasing time, $T_2^h$ in magnetic field is approximately 1 $\mu$s [3, 17].

We study here the emission from the $X^+$ state in a dot deterministically charged with a hole using a diode (Figure 1). We show photons from this transition display polarization correlation over a timescale one order of magnitude greater than the radiative lifetime when excited by a linearly polarized laser. This time is short relative to some reports of the hole spin polarization lifetime [14] and we show that this is a result of dephasing caused by the electron when the system is excited. We investigate the magnitude of the effect as a function of applied external magnetic field and radiative lifetime.

FIG. 1: (a) Schematic band-structure of the hole-charging diode when the dot contains a single hole. (b) Energy level diagram for the $X^+$ at zero magnetic field. (c) Spectrum under quasi-resonant excitation at 1.2 V. (d) Emission pattern for an $X^+$ transition showing $\beta = 0.092$.

The $X^+$ consists of two holes in a singlet $S = 0$ state and an electron. The zero net-hole-spin ensures the electron-hole anisotropic exchange interaction is absent. In zero magnetic field, the $X^+$ eigenstates are degenerate and labeled by the spin of the electron (Figure 1). $\beta$ [15, 16]. Radiative decay from $X^+$ to the single hole ground state occurs with a change in total angular momentum $\pm 1$, the polarization of the photon being corre-
lated with the initial and final spin state. In a dot where
the hole is “heavy” \((m_j = \pm 3/2)\) only the vertical transi-
tions in Figure 1(a) are allowed: the detection of a left-
handed photon (L) photon ensures the decay occurred by
the left hand transition on Figure 1(b).

The strain, shape anisotropy and inversion asymmetry in
InGaAs/GaAs QDs ensures the optically active transition has a mixed heavy- \((m_j = \pm 3/2)\) and light-
\((m_j = \pm 1/2)\) hole character [18, 19]. The state is given by 
\(\phi_{\pm} = (|\pm 3/2\rangle + |\beta \mp 1/2\rangle)/\sqrt{(1 + \beta^2)}\), which we
denote \(\uparrow\) and \(\downarrow\). Recombination of an electron and a
mixed heavy/light-hole now results in elliptically polar-
ized photons from \(X^+\), and \(\beta\) may be determined from
the emission pattern (Figure 1(iii) [18]). Within the sam-
ply studied \(\beta\) values from 0.02-0.20 are typical, and for the
data shown here \(\beta = 0.092\) (Figure 1(iii)).

![Figure 2](image)

FIG. 2: (a) Apparatus to measure the polarization corre-
lation from a dot. (b) Circular co- and cross- polarized emission
correlation from \(X^+\) at zero external field, \(g_{co}(t)\) (black) and
\(g_{cross}(t)\) (red), respectively. Extracted degree of polarization
correlation, \(C(t)\) (green). (c) The same measurement made
in the linear detection basis.

The diode for controlled charging has a 20 nm GaAs
tunnel barrier between the dot and p-contact (Figure 1(i)).
A 75% AlGaAs barrier on the n-side prevents electron
charging, so the \(X^+\) dominates at 1.2-1.3 V. Emission
from \(X^+\) at an energy of 1349.2 meV is excited quasi-
resonantly by a linearly polarized laser at 1317.2 meV. This
excitation scheme equally excites both transitions in
Figure 1 and the absence of spin pumping ensures there
is no build up of nuclear spin polarization, but it does
not populate other carrier combinations such as neutral
and negatively charged excitons (Figure 1(i)).

The experiment is shown in Figure 2. After filter-
ing, the emission is passed to polarization-maintaining
fibre-optics which enable 4 simultaneous measurements of
correlation in a basis selected by the quarter-wave plate
(QWP) and the polarizing coupler (PBS).

Figure 2 presents correlations recorded at zero exter-
mental field at an excitation power \(\times 10\) below saturation,
in the circular basis. Comparing the sum of the co-
and cross-polarized measurements \((g_{co}(t)\) in black and
\(g_{cross}(t)\) in red, respectively), we see a clear difference.
Note that both \(g_{co}(t)\) and \(g_{cross}(t)\) show a reduced signal
within \(\sim 1\) ns of zero-time delay due to the anti-bunched
nature of the light. Outside the central \(\sim 1\) ns there is an
enhanced probability of the source emitting two photons
of the same circular polarization over the case of emitting
photon of opposite circular polarization.

The degree of polarization correlation, \(C(t)\) is de-
finie as \(C(t) = (g_{co}(t) - g_{cross}(t))/(g_{co}(t) + g_{cross}(t))\)
from which a least squares fit with a function \(C(t) = \cos(-|t|/\tau_d)\) extracts the polarization correlation at
zero delay \(\tau_0\) and the timescale, \(\tau_d\). Empirically, this
function is a good fit to \(C(t)\) (Figure 2(a)). \(\tau_0 = 0.33\pm0.01\)
and the decay time of the correlation \(\tau_d = 9.0 \pm 0.4\) ns.
In contrast, measurements in the linear-polarization basis
\((H/V)\) show an absence of polarization correlation (Fig-
ure 2(a)).

Non-zero heavy-light hole mixing is an obvious source of
reduced polarization correlation. Taking the heavy:
light hole oscillator strength of 3:1 [18, 19] we see that
recombination of a \(\phi_+\), hole and an electron in the \(X^+\)
level leads to an elliptical photon with state \(\propto \sqrt{3}|L > +|\beta R >\), and a \(\phi_-\) hole. Conversely, decay involving a
\(\phi_-\) hole and an electron leads to a \(\propto \sqrt{3}|R > +|\beta L >\)
photon. The measurement in Fig. 2 is in the circular
basis so detection of a left-handed photon implies the de-
cay came from \(\phi_+\) with \(3/(3 + \beta^2)\) probability. In the
absence of dephasing in the upper or lower states, this
reduces the probability of obtaining sequential left-left
photo-detections to \((9 + \beta^4)/(3 + \beta^2)^2\). For this QD
\(\beta = 0.092\), so the probability of co-polarized photon emission
is reduced to 0.994. This is higher than we have mea-
sured, so we conclude an additional factor must be in-
cluded.

![Figure 3](image)

FIG. 3: A measurement of the timescale of correlation at zero
external magnetic field, \(\tau_d\), as a function of the normalized
intensity of the source. The data is fitted with an inverse
relationship.

In fact, the data can be explained by the fast dephas-
ing of the electron spin in the upper state, which dom-
inates any dephasing from the hole spin. A coincidence detection event arises as follows: the transition emits a photon that is detected with circular polarization and the hole spin is left in the corresponding state. Some time later, the system is re-excited to the upper state, where electron spin dephasing occurs during the radiative lifetime of the $X^+$ state, following which a second photon is emitted from the spontaneous decay. These two photons form a single coincidence in Figure 2(a). We stress that our model implicitly assumes the hole spin lifetime is greater than the measured $\tau_d$, though we envisage that future experiments that reduce the effect of electron dephasing it will be necessary to include the contribution of the hole.

Our studies provide four pieces of evidence electron spin dephasing is the factor limiting the polarization correlation. Firstly, the degree of polarization correlation from the $X^+$ observed in Figure 2(b) is 1/3. When excited, the unpaired electron spin evolves through a hyperfine interaction with the nuclei. Only those nuclear field fluctuations in the two directions perpendicular to the spin will cause precession. If this precession is faster than the radiative lifetime of the upper state, its effect is to randomize the spin. The electron spin parallel to the nuclear field is preserved. Thus, the mean spin projection along $z$ is reduced to 1/3. Secondly, the timescale over which polarization correlation is observed is inversely proportional to pump rate, as shown in Figure 3. This cannot be explained by dephasing occurring in the ground state. The increased excitation increases the number of times the system is excited between photon detection events, and this increases the rate at which the polarization correlation is lost. Thirdly, there is no polarization correlation in the linear basis (Figure 2(c)). This is consistent with a dephasing of the electron spin state in a time faster than the $X^+$ radiative lifetime. Finally, we shall show that change in $C_0$ with magnetic field is explained by the dynamics of the electron spin.

We next discuss the application of a Faraday magnetic field, which removes the degeneracy of the upper and lower states, shown in Figure 4. The net field experienced by the spins is the sum of the external field $B_{\text{ext}}$ and the internal nuclear field, $B_N$. This stabilizes the electron spin along $z$ and causes it to precess about the sum of the two fields, which is predominantly along the $z$ when $|B_{\text{ext}}| > B_N$. Thus, the application of vertical field increases the value of $C_0$ as shown in Figure 4(b). Figure 4 plots the polarization correlation timescale, $\tau_d$, versus magnetic field at constant laser intensity. This value changes from 9.0 ± 0.4 ns at zero field to 14.5 ± 0.5 ns at 300 mT.

A model of the dephasing of electron spin in QDs was presented by Merkolov, Efros and Rosen [20]. In this framework it is assumed that on timescales below 1 $\mu$s the hyperfine interaction between the electron spin and the nuclei in the dot can be considered semi-classically as a “frozen” magnetic field, of finite variance, but no directional preference. The electron $g$-factor is assumed isotropic. The time evolution of the electron spin $S(t)$, (initially along $S_0$) is given by:

$$S(t) = (S.n)n + (S_0 - (S_0.n)n)\cos(\omega t) + (S_0 - (S_0.n)n)\times n\sin(\omega t)$$

(1a)

$$W(B_N) \propto \exp\left(-\frac{(B_N)^2}{\delta B_N^2}\right)$$

(1b)

Where the distribution of nuclear field strengths, $W(B_N)$ is parameterized by the Gaussian width of fluctuations, $\delta B_N$. Figure 5a shows how the spin projection along the $z$ direction $S_z$ varies with the external magnetic field $B_{\text{ext}}$. At fields of a few times $\delta B_N$ the spin-projection along $z$ is stabilized. This has not eliminated the nuclear spin fluctuations, it has merely overwhelmed them at the cost of increased rate of precession about the net field. Any measurement along an orthogonal polarization direction will reveal the increased precession rate.

![FIG. 4](energy_level_diagram.png)  
(a) $B_{\text{ext}} > 0$, $X^+$  
(b) $g_+^+$  
(c) $g_0^+$

FIG. 4: Energy level diagram for the $X^+$ transition with finite $z$-magnetic field. (b) The degree of correlation, $C_0$ as a function of external Faraday magnetic field (black points). The calculated variation with field is shown as a red line. (c) Variation in the timescale of correlation, $\tau_d$.

From Equation 1a we extract the expected final electron spin projection along $z$, $S_z$, which is equal to the
polarization correlation $C_0$ (solid line in Figure 4b). The only fitting parameter is the width of the fluctuations in the nuclear-field $\delta B_N$, set to 100 mT. The dephasing time for this electron $\tau_0$ is therefore $T_\Delta = \hbar / g_e \mu_B \delta B_N \sim 200 \mu$s, which is, as expected, much less than the 1 ns radiative lifetime of the upper state. This provides a good fit to the data, reproducing the value of $C_0$ and width around zero field. The model fits less well at the higher fields. Partly, this can be explained by non-zero $\beta$, but the discrepancy requires further investigation.

The extracted $\delta B_N$ is within the range derived from a spin-noise measurement $\delta B_N$ but is greater than inferred from dephasing of the $X_0$ state in similar dots $\delta B_N$. We attribute this to the smaller wave-function extent of the electron when the dot additionally contains two holes. As $\delta B_N$ scales with $1/\sqrt{V}$ where $V$ is the volume of spins overlapping with the wave-function there is a variation in the effective $\delta B_N$ between states. This is the same reason the electron $g$-factor changes in the presence of additional holes $\delta B_N$.

To increase $C_0$ one could employ a host semiconductor without nuclear spin, a QD of greater volume or reduce the fluctuations in the nuclear field. Alternatively, Figure 5b shows that reducing $\tau_{rad}$ to the electron spin-lifetime leads to a significant increase in polarization correlation. This could be achieved by placing the dot into a cavity that equally enhances the radiative decay, independent of polarization.

In conclusion, despite the long hole spin coherence time in quantum dots the emission of polarization-correlated photons from the $X^+$ state is limited by electron spin dephasing. A significant increase in the polarization correlation time should be achieved by reducing the radiative lifetime of the $X^+$ state. Additionally, the degree of polarization correlation can be increased by applying an external magnetic field greater than the nuclear field.

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