**High-concurrence steady-state entanglement of two hole spins in a quantum dot molecular**

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Entanglement, a non-trivial phenomena manifested in composite quantum system, can be served as a new type of physical resource in the emerging technology of quantum information and quantum computation. However, a quantum entanglement is fragile to the environmental-induced decoherence. Here, we present a novel way to prepare a high-concurrence steady-state entanglement of two hole spins in a quantum dot molecular via optical pumping of trion levels. In this scheme, the spontaneous dispassion is used to induce and stabilize the entanglement with rapid rate. It is firstly shown that under certain conditions, two-qubit singlet state can be generated without requiring the state initialization. Then we study the effect of acoustic phonons and electron tunnelings on the scheme, and show that the concurrence of entangled state can be over 0.95 at temperature $T = 1K$.

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**Introduction**—Semiconductor technology toward quantum information science has opened up the possibility of constructing scalable quantum devices. As an attractive host for storing quantum information bit (qubit), electron or hole spins in self-assembled quantum dot (QD), are most promising for their scalability, relatively ease of coherent manipulations [1] and strong robustness against relaxation [2, 3]. In the past few years, significant theoretical and experimental works have been made towards controlling and entangling quantum dots. These experiments include the efficient state initialization [4] and coherent population trapping of single spin [2], the spin-readout [2] and single-spin Faraday/Kerr rotations for single quantum dot spin [2, 6], as well as the inter-dot coupling in double quantum dots molecules [7, 8]. Theoretically, various schemes for entangling quantum dots have also been proposed [2, 10].

However, all these schemes are exclusively tailored for electron spins in self-assembled quantum dot. Due to the longer coherence time compared with electron spin, hole spin in quantum dot has been paid more and more attention. A hole-spin state, which is constructed from a p-type atomic wave function, has many favorable aspects such as highly-suppressed hyperfine interaction [11] and much smaller tunneling rate [12], in comparison with electron spin. Recently both experiments have been reported for initializing single hole spin with high fidelity of 0.99 [11], and creating the coherent population trapping state [12].

In this letter, we present a scheme to generate high-fidelity steady state entanglement of two hole in a coupled QDM. Our scheme is based on spontaneous emission where the coupling between the QDM is dominated on Förster resonant. We show that our scheme is initial-state independent, and robust against decoherence and tunneling effect. At $T = 1K$, concurrence higher than 95% is possible in state-of-art technology.

**The model**—The QDM system composes two identity vertical aligned QDs, where an external magnetic field in the Voigt geometry [4] exists between such two resonant QDs. (b) and (c) Four level scheme illustrating the ground and excited states of a single self-assembled QD in the Voigt Configuration. (d)The preparation process of entangled state.

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| ∈ | +⟩, and the excited trion states are |⟩ = | ↑↓⟩ and |r⟩ = | ↑↑⟩, where ↑ (|⟩) and ↑ (|↓⟩) denote a heavy hole and an electron with spins along (against) x direction. As the magnetic field is along x direction, there is an additional Zeeman splitting $E_B = g_s^{\ast} \mu_B B x$ between ground (excited) states. Because of this splitting, four transitions between ground states and trion states can be independently addressed by polarization and frequency selection. Here we can choose $H = \sigma_+ - \sigma_-$ and $V = \sigma_+ + \sigma_-$.

Optical pumping protocol - In generally, the tunneling rate decreases exponentially when increasing the distance between two dots, while the Coulomb interaction, including both static dipole coupling $V_{xx}$ and Förster interaction $V_F$, decreases generally $d^{-3}$. So, it is possible to choose a property distance, where the Coulomb interaction dominant the interactions between the QDM. The Static dipole coupling $V_{xx}$ is the bi-trion energy shift, while the Förster interaction between the QDM is a kind of inter-dot interaction.

Due to the Voigt geometry magnetic field, the energy detuning between two $H$ type transitions are $\Delta H = E_{2}^g - E_{2}^g$, while the energy detuning between $V$ type transitions are $\Delta V = E_{2}^g - E_{2}^g$. In our scheme, we drive a $H$ polarized laser, and the detuning $H$ enables the transitions $H_1$ or $H_2$ to be independently addressed, which has been achieved in experiments with high fidelity[4]. In the following we choose the transition $H_1$, thus the energy levels of QDM employed in our scheme is described by three states: |0⟩, |1⟩ and |s⟩ (Fig.1(c)). The frequency and Rabi frequency with $H$ polarized laser are supposed to be $\omega$ and $\Omega$ respectively. Additionally, since the direct excitation of the transition |0⟩ ↔ |1⟩ is forbidden, $\Omega_m$ can be realized by employing a Raman transition with large detuning to an auxiliary excited state. The Hamiltonian of the QDM reads

$$H = \sum_{i=1,2} \left[ (\Omega_1)|i⟩⟨1| + \Omega_m|0⟩⟨1| + H.c. \right] + \omega|s⟩⟨s| + V_F(|1⟩⟨s| + |s⟩⟨1|) + V_{xx}|ss⟩⟨ss|.$$  

Since Hamiltonian Eq. (1) is of the symmetry formation, it is convenient to introduce symmetric state $|S_{ij}\rangle = \frac{1}{\sqrt{2}}(|ij⟩ + |ji⟩)$ and anti-symmetric state $|A_{ij}\rangle = \frac{1}{\sqrt{2}}(|ij⟩ - |ji⟩)$ ($i,j = 0,1,s$). With the aid of Förster interaction, an energy shift is generated between symmetric excited states $|S_{01}\rangle$, $|S_{1s}\rangle$ and anti-symmetric excited states $|A_{0s}\rangle$, $|A_{1s}\rangle$.

When the $H$ polarized laser is driven to pump $H_1$ transition with detuning $\Delta = -V_F$, the transitions $|S_{01}\rangle ↔ |S_{0s}\rangle$ and $|11⟩ ↔ |S_{1s}\rangle$ are resonant in the rotating frame. In the case $\Omega_\infty \ll |V_F| \ll V_{xx}$, the populations on bi-trion and anti-symmetric single-trion states are nearly equal to zero, and can be eliminated adiabatically. Using the symmetric and anti-symmetric notation we introduced above, the scheme is reduced to a 6-state system (Fig.4). The effective Hamiltonian can be written as

$$H_{\text{eff}} = \sqrt{2}\Omega|1⟩⟨S_{1s}| + \Omega|S_{01}\rangle⟨S_{0s}| + \Omega_m|S_{0s}\rangle⟨S_{1s}|$$

$$+ \sqrt{2}\Omega_m|0⟩⟨S_{01}| + \sqrt{2}\Omega_m|S_{01}\rangle⟨S_{1s}| + h.c.$$  

Then we take the lifetime of trion states into account. The photon emission occurs via an decay of the state |s⟩ into |1⟩ with $\Gamma_1$ or into |0⟩ with $\Gamma_0$. The total spontaneous rate is assumed to be $\Gamma = \Gamma_0 + \Gamma_1$. Then we derive a master equation within a Markovian process,

$$\dot{\rho} = -i[H_{\text{eff}}, \rho] + \sum_i \left( 2\Omega|l_i⟩⟨l_i| + \frac{1}{2} (L^\dagger L)_{+} \right),$$  

where $L_1 = \sqrt{2}\Omega|00⟩⟨S_{0s}| + \frac{1}{\sqrt{2}}|S_{01}\rangle⟨S_{1s}|$, $L_2 = -\sqrt{2}\Omega/2|A_{01}\rangle⟨S_{1s}|$, $L_3 = \sqrt{2}\Omega|11⟩⟨S_{1s}| + \frac{1}{\sqrt{2}}|S_{01}\rangle⟨S_{0s}|$, $L_4 = \sqrt{2}\Omega/2|A_{01}\rangle⟨S_{0s}|$. The basic preparation cycle in our scheme as follows. The lasers with Rabi frequency $\sqrt{2}\Omega_M$ produce the transitions |00⟩ ↔ |S_{01}| ↔ |11⟩. Attribute to the Förster interaction, energy splitting generates between the symmetric and anti-symmetric states. By tuning the laser frequency to resonant with the symmetric single-trion states, the transitions $|S_{01}\rangle ↔ |S_{0s}\rangle$ and $|11⟩ ↔ |S_{1s}\rangle$ are created. Then the lasers couple the three ground states |00⟩, |S_{01}| and |11⟩ to the single-trion states |S_{0s}| and |S_{1s}|, and leave the entangled state $|A_{01}\rangle = \sqrt{2}|(10) - (01)|$ decoupled from them. On the other hand, spontaneous radiation performs from |s⟩ to |0⟩ and |1⟩, which is dissipation from single-trion states to subspace $M_1 = \{|00⟩, |S_{01}|, |11⟩\}$ and $M_2 = \{|A_{01}\}|$. If the excited states decay into $M_2$, the process is terminated when one trion is dissipated; if they decay into $M_1$, it will go through the cycle again. Therefore, after a period of time, the trions are dissipated and the system will go into a steady state $|A_{01}⟩$, which is the maximum entangled state we require.

For a potential experimental system, we can choose a QDM including two resonant QDs. The wave functions for electrons and holes in each QD is supposed to have a Gaussian form $\phi_{z/h} \sim \exp[-(x^2 + y^2 + z^2)/l_z^2/l_h^2]$, where the characteristic lengths $l_z = 4.4$ nm, $l_h = 4$ nm, $l_x = 1$ nm. The Förster interaction $V_F$ read as $|V_F| = \frac{\epsilon}{4\pi\varepsilon_0\epsilon_r}(\frac{l_z^2}{l_h^2})^2 F(x)$, where $\epsilon$ is the dielectric constant, $d$ is the distance between the QDM, $\lambda^2 = 2/(1/l_z^2 + 1/l_h^2)$ and $a \sim h/\sqrt{2m_0l_xl_{\epsilon_{\infty}}}$. In the case $l_zl_h \gg l_x$, $F(x) = \frac{x^3}{2} \int_0^\infty dl_z \frac{1}{l_z^2} \exp[-v]^2$, where $v = \frac{x^2}{l_z^2}$. In our scheme, we set the distance between two dots in QDM is $9.5$ nm and $|a| = 1.6$ nm. We can estimate $V_F = -0.2$ meV and $V_{xx} = 3$ meV. If the $x$-direction magnetic field is $B_z = 1$ T, and g-factors are $g_h = -0.29$ and $g_e = -0.46$, the Zeeman splitting of trion and ground states are about
to the requirement of frequency selection, the Rabi fre-
quency is fixed, the character time can be shorten as
expresses that the time for achieving the steady state
\[ T_0 \]
we consider is given by
\[ \rho_{T} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \]
illustrated in Fig.2(b) that the character time
\[ 1 \]
suppose that \( \Gamma \) with spontaneous radiation, which shows a near unity
|µeV| is suitable.
\[ B = eV \]
Fig.2(a) shows the entanglement of the two hole spins
in steady state measured by concurrence \( C \) as a function of time[19]. The Red solid line represents the case only with spontaneous radiation, which shows a near unity concurrence at \( T = 0 \) K after a period of time. Here we suppose that \( \Gamma_1 = \Gamma_2 = \Gamma/2 \). The input density matrix we consider is given by \( \rho_{i} = \frac{1}{2}(|00\rangle\langle 00| + |S_{01}\rangle\langle S_{01}| + |A_{01}\rangle\langle A_{01}| + |11\rangle\langle 11|) \). After a period of time, the output state ideally generates as \( |\Phi_{f}\rangle = |A_{01}\rangle \). If the evolution time \( t \to \infty \), the density matrix \( \rho \to |A_{01}\rangle\langle A_{01}|. \) It is illustrated in Fig2(b) that the character time \( T_0 \), which expresses that the time for achieving the steady state[20],
depends on spontaneous radiation rate \( \Gamma \) and the Rabi frequency of pumping field \( \Omega \). If the rate of spontaneous emission is fixed, the character time can be shorten as
Rabi frequency of laser field increases, and finally saturates approximal to a value \( T_0 \sim 10/\Gamma \), which is ten times that of the lifetime of trion state. The optimal characteristic time is appeared when we tune the coupling \( \Omega_m \) to satisfy \( \Omega_m = 0.45\Omega \)(shown in Fig2(b)).

For a real QDM, the rate of the spontaneous radiation from trion state to electronic state is about \( 1.2 \) µeV, therefore \( T_0 \) has a value about \( 5.5 \) ns.

The effect of phonon interaction - In Fig1, we have taken exciton as the auxiliary state, which is vulnerable by the vibrational modes of the surrounding phonons. The interaction between acoustic phonons and excitons may be mediated by deformation potential coupling and piezoelectric coupling. Thus the phonon coupling matrix element[21] is
\[ g_{q,i} = e^{i\mathbf{q}\cdot\mathbf{d}_i}[\hat{M}_{q,i}^e(q) - \hat{M}_{q,i}^p(q)], \]
where \( \hat{M}_{q,i}^e(q) = \sum_{\mathbf{q},\mathbf{i}} \sqrt{\frac{\hbar}{2\mu_{q,i}}} (|q|D_{e(h)} + i\mathbf{p}_q), \rho_{e/h}(q) = \int d^3r |\phi_{e/h}\rangle e^{i\mathbf{q}\cdot\mathbf{r}}. \) Following the Markovian approximation[22], the master equation of the density matrix in the interaction picture with respect to \( H \) may be reduced into a Lindblad form
\[ \dot{\rho} = \sum_i J(\omega_i)[(N_i + 1)D[P_i]\rho + N_iD[P_i^\dag]\rho], \]
where \( D[P]\rho = P\rho P^\dagger - \frac{1}{2}\{P^\dagger P\} \) is the decay operator of phonon effect, and \( N_i = [\exp(\omega_i/k_BT) - 1]^{-1}. \) \( J(\omega_i) \) denotes the phonon spectral density, and there are two kinds of \( J(\omega_i) \) in our model which can be written as
\[ J_{\pm}(\omega) = \int d\Omega(1 \pm \text{sinc}(\frac{\omega d}{\Omega}))|\mathcal{G}_d(\omega) + \mathcal{G}_p(\omega)|, \]
where \( \mathcal{G}_d(\omega) = \frac{\omega_d^2}{8\pi^2\mu_c^2}(D_q\rho_e - D_h\rho_h)^2, \mathcal{G}_p(\omega) = \frac{e^2}{8\pi^2\mu_c^2}(\rho_e - \rho_h)^2. \) Here the piezoelectric coupling is \( P_q = \frac{1}{4}\sin\theta M_p\sqrt{9 + 7\cos2\theta - 2\cos4\theta}\sin^2\theta, \) in which \( M_p \) denotes piezoelectric constant[23]. Combining the phonon effect into the Master equation Eq. (5), we will get numerical results of the concurrence shown in Fig2(b). As temperature increases, the concurrence of entangled state decreases. The parameters for the phonons is taken from Ref. [21].

The effect of tunneling effect - The tunneling effect is assumed to be much smaller than the Förster interaction[24] in above discussion. This assumption can be technically accomplished by increasing the distance between double dots but simultaneously the Förster interaction is suppressed. It is demonstrated that the tunneling effect of hole is less than that of electron by one or two orders of magnitude[12] which guarantees the stability of the hole-included ground state. Thus, we primarily consider the influence of electron tunneling on our scheme. Using standard WKB method, the tunneling rate can be estimated to \( t_e \approx \frac{2e}{\pi}\sqrt{8\hbar\omega}\exp[-\frac{16V_F}{3\omega}], \) with
tron(hole) generates in the \( i \) as exciton(an electron-hole pair lies in the same dot), such that tunneling effect, the trion might include inter-dot excitation caused by inter-dot exciton. Therefore the effect as basis as tunneling effect into an interaction picture with respect to \( \theta \), and \( \sin \theta \) for different electron tunneling rates: (a) as a function of time \( \omega = \sqrt{2}V_e/d \). If the case is \( t_e = 1.9 \) meV, which can not be omitted compared to the Förster coupling \( |V_F| = 0.2 \) meV.

The exciton we discuss above in trion \( |s\rangle \) is intra-dot exciton(an electron-hole pair lies in the same dot), such as \( |e_1^ih_1^i\rangle \) or \( |e_2^ih_2^i\rangle \), where \( e_i^i(h_i^i) \) denotes that one electron(hole) generates in the \( i \)th dot. Due to the electron tunneling effect, the trion might include inter-dot exciton \( (|e_1^ih_2^i\rangle \text{ or } |e_2^ih_1^i\rangle) \), and we denote this kind of trion as \( |t\rangle \). Here, we do not consider the spontaneous radiation caused by inter-dot exciton. Therefore the effect of inter-exciton gives a new contribution to the initial Hamiltonian describing by Eq. (1) as

\[
H_1 = \sum_{i=1,2} (\omega_i|t\rangle_i\langle t| + t_c(|s\rangle_i\langle t| + h.c.)).
\]

We move the combined hamiltonian including tunneling effect into an interaction picture with respect to \( \sum_{i=1,2} (\omega+V_F)|s\rangle_i\langle s| + |t\rangle_i\langle t| \), and proceed by transforming the single exciton part of hamiltonian into a new basis as \( |\psi_1(3)\rangle = \cos\theta|S_{0(1)}(3)\rangle - \sin\theta|S_{0(1)}(1)\rangle \), \( |\psi_2(4)\rangle = \sin\theta|S_{0(1)}(3)\rangle + \cos\theta|S_{0(1)(1)}\rangle \), where \( \theta = -\frac{1}{2}\arccot(\frac{\delta}{\omega}) \)

and the detuning \( \delta = V_F + \omega - \omega_i \). These two eigen states \( |\psi_1\rangle \) and \( |\psi_3\rangle \) are degenerated at energy \( E_1 = \frac{1}{2}(\delta + \sqrt{4\delta^2 + \omega^2}) \), while \( |\psi_2\rangle \) and \( |\psi_4\rangle \) are degenerated at energy \( E_2 = \frac{1}{2}(\delta - \sqrt{4\delta^2 + \omega^2}) \). If we select the frequency of pumping laser as \( \omega_i = \omega + V_F + E_1 \), and guarantee the condition \( \Omega \ll |E_1 - E_2| \), the effective Hamiltonian becomes as

\[
H_{\text{eff}} = \Omega_m(\sqrt{2}|00\rangle\langle S_{01}| + \sqrt{2}|S_{01}\rangle\langle 11| + |S_{0k}\rangle\langle S_{1s}|) \\
+ \Omega \cos \theta(\sqrt{2}|11\rangle\langle \psi_3| + |S_{01}\rangle\langle \psi_1|) + h.c. \quad (8)
\]

We consider the influence of both tunneling effect and phonon interaction and follow similar method as in Eq. (4). The concurrence of hole spin entangled state as a function of time at \( T = 1K \) is shown in Fig. 3(a). We can find that the concurrence is beyond 95%, and the phonon-exciton process might be suppressed by decreasing electron tunneling rate. When the electron tunneling rate is slow as \( t_e \ll |\omega - \omega_i| \), the Eq. (8) can be reduced to Eq. (2). It indicates that the small electron tunneling does nothing more than an energy shift of the single exciton states whose effect can be offset by tuning the Rabi frequency of the external laser field. Fig. 3(b) illustrates the concurrence of the steady state as a function of experimental temperature \( T \). The high concurrence of stationary entangled state is much less influenced when decreasing the temperature, and the concurrence is more susceptive to temperature if electron tunneling rate increases. In our scheme, the electron tunneling has a value of \( t_e \sim 2 \) meV, which is much smaller than the energy gap between inter-exciton and intra-exciton(\( \sim 20 \) meV). Thus, the hole spin entangled state remains robust even if electron tunneling effect is considered.

Conclusion - To sum up, we have shown that a stationary entangled state on spins with high concurrence can be prepared in a quantum dot molecular by technically designing the spontaneous dispassion processes. The hole spin for its small inter-dot tunneling rate is more suitable to encode qubit compared with electron spin in our scheme. We also discuss the influence of phonon-exciton interaction and electron tunneling effect on the entangled state. For the real experiment with \( t_e = 2 \) meV, the concurrence of entangled state is still over 95% at \( T = 1K \).

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