

# Theory of electron–hole exchange interaction in double quantum dots

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A theory of electron–hole exchange interaction in vertically stacked double quantum dots is developed. This theory was built up from the full interacting Hamiltonian of two coupled dots, taking into account both of the macroscopic and microscopic natures of particle wave functions. In the basis of S-type Fock–Darwin orbitals, compact formulations for electron–hole exchange terms responsible for the fine struc-

ture splitting of spin exciton states as functions of material parameters, geometric structure of dots, and the strength of external field are explicitly derived. Significant reduction of the splitting, down to 50% of the magnitude of such energy in single dot cases, is observed as the system is switched into the near-resonance regime.

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**1 Introduction** Self-assembled quantum dots (QDs) have been proposed as potential sources in entangled photon pairs production. However, a main obstacle to successful generation of entangled photon pairs from QDs is the fine structure splitting (FSS) between intermediate one-exciton (X) spin states [1, 2], a direct consequence of the electron–hole (e–h) exchange interaction (EI) intrinsically existing in solids [3]. In this work we develop a microscopic theory of electron–hole exchange interactions and excitonic fine structure in double quantum dots (DQDs). We derive the explicit expressions for the e–h EI between the direct-direct (DD), direct-indirect (DI) and indirect-indirect (II) excitons in DQDs. The developed approach is used to evaluate the dependence of FSS for DQD systems on the unique tuning parameters such as bias field and the interdot distance. With the advantage of extra tuning parameters, DQD systems are shown to provide a feasible way for controlling the FSS of exciton mediating states and entanglement in the emitted photons.

In the next section, we start with the generalized Hamiltonian of interacting e–h pairs in coupled dots, and then show an explicit expression for the e–h EI part determined by the complete Bloch wave functions. Next to that, we use Fock–Darwin (F–D) orbitals as basis for expansion of one-exciton spin states and derive the explicit expressions

for direct and indirect excitonic states. Within the chosen basis, we carry out numerical diagonalization for the corresponding Hamiltonian matrix, and analyze the calculated energy and polarized photoluminescence (PL) spectrum for a typical double dot system. We conclude in the last section.

**2 Model Hamiltonian** The full interacting e–h Hamiltonian for a coupled DQD system can be expressed, within the tight binding scheme, as

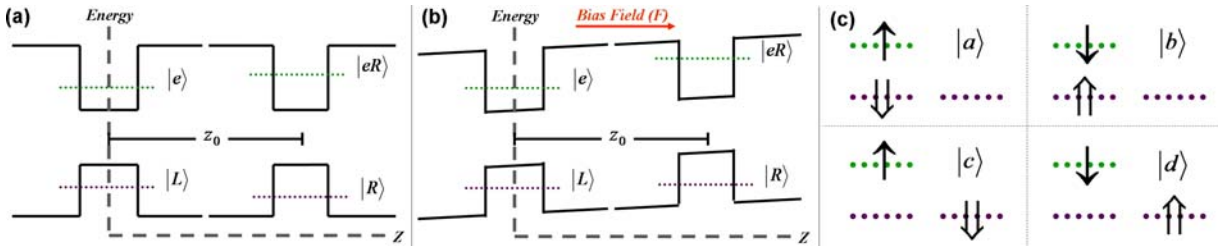
$$\hat{H} = \hat{H}^{ke} + \hat{H}^{kh} + \hat{H}^{eT} + \hat{H}^{hT} + \hat{H}^{ee} + \hat{H}^{hh} + \hat{H}^{eh} + \hat{H}^{eh-EI}, \quad (1)$$

where the term

$$\hat{H}^{ke} = \sum_j \epsilon_c^j c_j^\dagger c_j \left( \hat{H}^{kh} = \sum_n \epsilon_h^n h_n^\dagger h_n \right)$$

measures the kinetic energies of electrons (holes),

$$\hat{H}^{eT} = \sum_{jk} t_e^{jk} (c_j^\dagger c_k + \text{c.c.}) \left( \hat{H}^{hT} = \sum_{nm} t_h^{nm} (h_n^\dagger h_m + \text{c.c.}) \right)$$



**Figure 1** Schemes of the S-like isolated-dot orbitals of double dot systems (a) without bias field and (b) with positive applied bias field. (c) Configurations of spin bright X.

describes the particle hopping between adjacent dots in terms of the hopping parameters  $t_e^{jk}$  and  $t_h^{nm}$ ,

$$\hat{H}^{ee} = \sum_{jkk',\sigma\sigma'} V_{ee}^{jkk'} c_{j,\sigma}^\dagger c_{k,\sigma'}^\dagger c_{k',\sigma} c_{j',\sigma},$$

$$\hat{H}^{hh} = \sum_{mmn',\chi\chi'} V_{hh}^{mmn'} h_{n,\chi}^\dagger h_{m,\chi'}^\dagger h_{m',\chi} h_{n',\chi'}$$

and

$$\hat{H}^{eh} = - \sum_{njkm,\sigma\chi} V_{eh}^{njkm} h_{n,\chi}^\dagger c_{j,\sigma}^\dagger c_{k,\sigma} h_{m,\chi}$$

are the conventional e–e, h–h and e–h Coulomb interactions, where symbols  $j, k$  ( $n, m$ ) are respectively preserved throughout this article for labeling single electron (hole) states of an isolated dot as composite indexes containing the information about orbitals and position of dot (L or R),  $\sigma = \uparrow / \downarrow$  ( $\chi = \uparrow / \downarrow$ ) denotes electron (hole) spin (pseudospin), and  $c_{j\sigma}^\dagger$  and  $c_{j\sigma}$  ( $h_{n\chi}^\dagger$  and  $h_{n\chi}$ ) are the electron (hole) creation and annihilation operators, respectively.

The Coulomb matrix element defined by

$$V_{jkk'}^{ee} \equiv \iint d^3 r_1 d^3 r_2 \Phi_j^{e*}(\mathbf{r}_2) \Phi_k^e(\mathbf{r}_1) \times \frac{e^2}{4\pi\epsilon|r_1 - r_2|} \Phi_k^e(\mathbf{r}_1) \Phi_j^e(\mathbf{r}_2),$$

describes the two-particle Coulomb scattering process from initial states  $|k\rangle$  and  $|j\rangle$  to some final states  $|k'\rangle$  and  $|j'\rangle$ , in which conservation of particle spin is ensured, where  $\Phi_k^e$  ( $\Phi_n^h$ ) is the electron (hole) envelope wave function of the initial (final) state and  $\epsilon$  is the dielectric constant of the dots material [4]. The last term in Eq. (1) represents the e–h exchange interaction

$$\hat{H}^{ehEI} = - \sum_{njkm,\sigma\sigma'\chi\chi'} \delta_{eh}^{nj,\sigma,k\sigma',m\chi'} h_{n,\chi}^\dagger c_{j,\sigma}^\dagger c_{k,\sigma'} h_{m,\chi'}$$

in terms of the matrix elements defined by

$$\delta_{eh}^{nj,\sigma,k\sigma',m\chi'} = \iint d^3 r_1 d^3 r_2 \Phi_n^{h*}(\mathbf{r}_1) u_{v\chi}^*(\mathbf{r}_1) \times \Phi_j^{e*}(\mathbf{r}_2) u_{c\sigma}^*(\mathbf{r}_2) \frac{e^2}{4\pi\epsilon|r_1 - r_2|} \times \Phi_k^e(\mathbf{r}_1) u_{c\sigma'}(\mathbf{r}_1) \Phi_m^h(\mathbf{r}_2) u_{v\chi'}(\mathbf{r}_2), \quad (2)$$

where  $u_{c\sigma}$  ( $u_{v\chi}$ ) is the periodic part of electron (hole) Bloch function at the conduction (valence) band with spin  $\sigma$  ( $\chi$ ). Notable is that the spins of an electron and a hole could be simultaneously flipped in the Coulomb scattering via the e–h EI, leading to the lifting of the degeneracy of spin exciton states and being the main target of this work. Since our interest is in bright exciton states, here we consider the following four spin configurations of bright exciton,  $\delta_{eh}^{n\downarrow,j\uparrow,k\uparrow,m\downarrow} = \delta_{eh}^{n\uparrow,j\downarrow,k\downarrow,m\uparrow} = \delta_0^{njkm}$  and  $\delta_{eh}^{n\downarrow,j\uparrow,k\downarrow,m\uparrow} = \delta_{eh}^{n\uparrow,j\downarrow,k\uparrow,m\downarrow} = \delta_1^{njkm}$ ; the former one with the typical energy value at the scale  $\delta_0^{njkm} \sim 10^2 \mu\text{eV}$  is responsible for the splitting of bright and dark excitons, while the latter with  $\delta_1^{njkm} \sim 10^1 \mu\text{eV}$  for the mixing of bright X with opposite spins and the corresponding FSS [5]. On the other hand, the integral in Eq. (2) is customarily decomposed into two parts ( $\delta_{eh}^{nj,\sigma,k\sigma',m\chi'} = \delta_{eh-Sr}^{nj,\sigma,k\sigma',m\chi'} + \delta_{eh-Lr}^{nj,\sigma,k\sigma',m\chi'}$ ), i.e. the short range (Sr.) part for  $|r_1 - r_2| < a_0$  and the long range (Lr.) one for  $|r_1 - r_2| > a_0$ , where  $a_0$  is the lattice constant. It is known that the main contribution to  $\delta_1^{njkm}$  comes from the Lr. part [6]. Then, within the dipole-dipole approximation this term is finally written as [4]

$$\delta_1^{njkm} \approx \delta_{eh-Lr}^{nj,\sigma,k\chi,m\sigma} = \delta_{\sigma,\chi'} \delta_{\chi,\sigma'} \delta_{\sigma,-\sigma'} \frac{3e^2 \hbar^2 E_p}{2\epsilon m_0 E_g} \iint d^3 R d^3 \Delta R \times \Phi_n^{h*}(\mathbf{R}) \Phi_j^{e*}(\mathbf{R} - \Delta\mathbf{R}) \Phi_k^e(\mathbf{R}) \Phi_m^h(\mathbf{R} - \Delta\mathbf{R}) \times \frac{(\Delta R_y^2 - \Delta R_x^2) + 2i\Delta R_x \Delta R_y}{|\Delta\mathbf{R}|^5}, \quad (3)$$

where we have renamed variables  $R \equiv r_1$  and  $\Delta R \equiv r_1 - r_2$ , making easier of noting the Lr. character of the term. We limit our study to a single neutral exciton confined in a double quantum dot system with interdot distance  $z_0$  and applied bias field, as depicted in Figs. 1(a) and (b). Focusing on coupled quantum dots under low-power excitation conditions and at low temperature, we work on single X states involving the main three S-type orbitals of isolated-dots [7]. Here we adopt this three-orbital model for the calculation of excitonic spectrum of bias-controlled DQD, regarding it has been successfully applied to the explanation of previous experimental results [8].

Based on the single particle states shown in Fig. 1(a), the four relevant single exciton configurations are built up as follows,

$$|a(b)\rangle \equiv |L\rangle |\downarrow(\uparrow)\rangle |e\rangle |\uparrow(\downarrow)\rangle = h_{L,\downarrow(\uparrow)}^\dagger c_{e,\uparrow(\downarrow)}^\dagger |0\rangle, \quad (4a)$$

$$|c(d)\rangle \equiv |R\rangle |\downarrow(\uparrow)\rangle |e\rangle |\uparrow(\downarrow)\rangle = h_{R,\downarrow(\uparrow)}^\dagger c_{e,\uparrow(\downarrow)}^\dagger |0\rangle. \quad (4b)$$

Figure 1(c) shows the chosen single particle orbital basis and bright spin-exciton configurations. Configurations  $|a\rangle$  and  $|b\rangle$  are related to direct X while  $|c\rangle$  and  $|d\rangle$  to indirect X. Since our interest is in the fine structure of bright excitons, we reduce the terms apparently irrelevant to the FSS by despising Coulomb interaction and non spin flip e-h EI for indirect states ( $V_{eh}^{ReeR} = \delta_0^{ReeR} = 0$ ) and by removing the common offset of single particle energy and non spin flip e-h EI for direct states to PL spectra of single excitons ( $\varepsilon_e^c = \varepsilon_h^L = \varepsilon_h^R = \delta_0^{LeeL} = 0$ ). We further rename the variables,  $V_{eh}^{LeeL} = V_{eh}$ ,  $t_h^{LR} = t_h$ ,  $\delta_1^{LeeL} = \delta^{DD}$ ,  $\delta_1^{ReeL} = \delta^{DI}$  and  $\delta_1^{ReeR} = \delta^{II}$ , for brevity of expressions. It turns out that the Hamiltonian can be written in a compact form

$$\hat{H} = \begin{pmatrix} -V_{eh} & -\delta^{DD} & -t_h & -\delta^{DI} \\ -\delta^{DD} & -V_{eh} & -\delta^{DI} & -t_h \\ -t_h & -\delta^{DI} & -eFz_0 & -\delta^{II} \\ -\delta^{DI} & -t_h & -\delta^{II} & -eFz_0 \end{pmatrix}, \quad (5)$$

where  $e$  is the elementary charge,  $F$  is the bias field, and the off-diagonal terms  $\delta^{DD}$ ,  $\delta^{DI}$ , and  $\delta^{II}$  couple the spins of DD, DI and II pairs of exciton states, respectively. After diagonalization we obtain the energy spectrum of single spin exciton in DQDs versus bias field, exhibiting two anticrossed energy branches (denoted by  $E_1$  and  $E_2$  for the lower branch, and  $E_3$  and  $E_4$  for the upper one) with the bias-controlled FSS, as shown in Fig. 2(a). From existing experiments [6] and Eq. (3), one deduces that  $|\delta^{DI}| \ll |t_h|$ , and  $\delta^{II} < \delta^{DI} < \delta^{DD} \ll V_{eh}$ , and by utilizing the relationship of energy scale differences, we derive the FSSs ( $E_2 - E_1 \equiv \Delta E_{low}(F, z_0)$  and  $E_4 - E_3 \equiv \Delta E_{up}(F, z_0)$ ) as functions of bias field and inter-dot distance

$$\Delta E_{up/low}(F, z_0) \approx \delta^{DD} + \delta^{II} \pm T(F, z_0), \quad (6)$$

with

$$T(F, z_0) = \frac{(\delta^{DD} - \delta^{II})(eFz_0 - V_{eh}) - 4\delta^{DI}t_h}{\sqrt{4(t_h)^2 + (eFz_0 - V_{eh})^2}}.$$

From these expressions, it is clear that the FSSs approach the values  $\Delta E_{low}(F, z_0) \rightarrow 2\delta^{DD}$  and  $\Delta E_{up}(F, z_0) \rightarrow 2\delta^{II}$  in low field limit  $F \rightarrow 0$ , and  $\Delta E_{low}(F, z_0) \rightarrow 2\delta^{II}$  and  $\Delta E_{up}(F, z_0) \rightarrow 2\delta^{DD}$  in high field limit ( $F \rightarrow \infty$ ). In turn, FSSs are close to the values  $\Delta E_{low}(F, z_0) \rightarrow \delta^{DD} + \delta^{II} + 2\delta^{DI}$  and  $\Delta E_{up}(F, z_0) \rightarrow \delta^{DD} + \delta^{II} - 2\delta^{DI}$  as DQDs are brought by field into the states near resonance ( $F \rightarrow V_{eh}/z_0$ ). This indicates that significant change of FSS might be achieved by means of bias control within the explicit interval  $\{2\delta^{II}, 2\delta^{DD}\}$ .

**3 Fock-Darwin orbital basis** Now to model the single particle states of QD we use the F-D states for the 3D parabolic potential. These envelope wave functions are explicitly given by

$$\Phi_e^c(\mathbf{R}) = k_e e^{-\frac{1}{2}\left(\left(\frac{x}{l_x}\right)^2 + \left(\frac{y}{l_y}\right)^2 + \left(\frac{z}{l_z}\right)^2\right)}, \quad (7a)$$

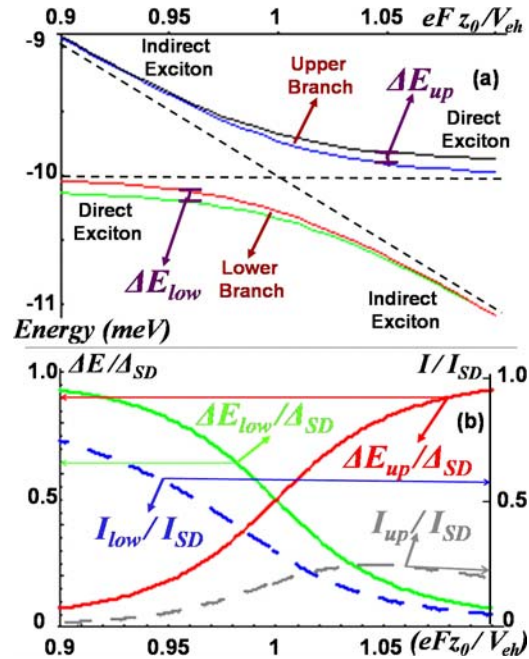
$$\Phi_L^h(\mathbf{R}) = k_h e^{-\frac{1}{2}\left(\left(\frac{x}{l_x}\right)^2 + \left(\frac{y}{l_y}\right)^2 + \left(\frac{z}{l_z}\right)^2\right)}, \quad (7b)$$

$$\Phi_R^h(\mathbf{R}) = k_h e^{-\frac{1}{2}\left(\left(\frac{x}{l_x}\right)^2 + \left(\frac{y}{l_y}\right)^2 + \left(\frac{z-z_0}{l_z}\right)^2\right)}, \quad (7c)$$

where  $k_e$  and  $k_h$  are the normalization constant of electron and hole wave functions, and  $l_x$  ( $l_y$  and  $l_z$ ) is the characteristic length measuring the electron wave function extension along the  $x$  ( $y$  and  $z$ ) direction. Substituting Eq. (7) in Eq. (3) and performing the integration, we obtain the following formulation of the e-h exchange interaction terms for slightly deformed dots,

$$\delta^{DD} = \delta^{(M)} \frac{(l_x - l_y)}{l_y^3}, \quad (8a)$$

$$\delta^{DI} = \delta^{DD} e^{-\frac{z_0^2}{4l_z^2}} \quad \text{and} \quad \delta^{II} = \delta^{DD} e^{-\frac{z_0^2}{l_z^2}}, \quad (8b)$$



**Figure 2** (online colour at: www.pss-b.com) (a) Energy spectrum of first four bright states from a double quantum dot as function of the normalized bias field (solid lines). Dashed lines indicate uncoupled direct and indirect X energies as reference. Green line:  $E_1$ , red line:  $E_2$ , blue line:  $E_3$  and gray line:  $E_4$ . (b) Fine structure splitting of the bright exciton states and the intensity of the corresponding polarized PL versus the applied bias fields. (Intensity normalized to the corresponding magnitudes of the single dot case  $I_{SD}$ .) Green line:  $\Delta E_{low}/2\delta^{DD}$ , red line:  $\Delta E_{up}/2\delta^{DD}$ , blue line:  $I_{low}/I_{SD}$  and gray line:  $I_{up}/I_{SD}$ .

where  $\delta^{(M)} = 3e^2 \hbar^2 E_p / 8\epsilon m_0 E_g^2 a_0$  is a constant associated with material properties. From these expressions, it is clear that finite value of  $\delta^{DD}$  arises from the lateral deformation ( $l_x \neq l_y$ ) of dot shape while that of  $\delta^{DI}$  and  $\delta^{II}$  involve both, the lateral deformation and inter-dot distance. Figure 2(a) shows the energy spectrum of single spin exciton in a DQD with  $z_0 = 10$  nm. Magnitudes of FSSs of the corresponding energy branches are presented in Fig. 2(b). The geometrical parameters  $l_x = 10/\sqrt{2}$  nm,  $l_x/l_y = 0.97$  and  $l_z = 1.5/\sqrt{2}$  nm are used here. We calculated for  $\text{In}_{0.5}\text{Ga}_{0.5}\text{Al}/\text{GaAs}$  DQDs with material parameters following Refs. [4] and [9]. With such numerical values for the system and  $t_h = 0.3$  meV, we obtain  $\delta^{DD} = 54$   $\mu\text{eV}$  and  $\delta^{DI} \approx \delta^{II} \approx 0$ .

**4 Polarized photoluminescence spectrum** The polarized PL spectra from single spin excitons in DQDs are calculated using Fermi's golden rule

$$I_{x(y)} \propto \sum_i f(E_i, T) |\langle 0 | P_{x(y)}^- | i \rangle|^2 \delta(E_i - \hbar\omega),$$

where  $i$  ( $f$ ) denotes initial (final) states of recombination transition and

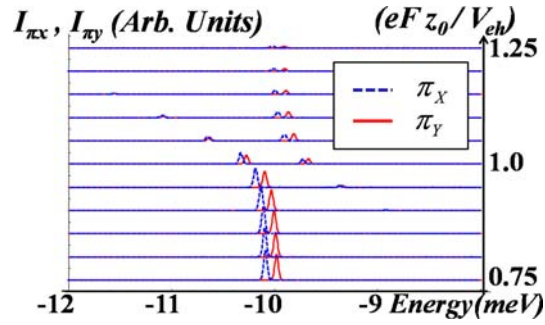
$$P_{x(y)}^{(-)} = \frac{1}{\sqrt{2}(i\sqrt{2})} \sum_{n,j} S_{n,j} (h_{n\uparrow} c_{j\downarrow} + (-) h_{n\downarrow} c_{j\uparrow}).$$

Considering single exciton eigenstates from diagonalizing the Hamiltonian in Eq. (6) as the transition initial states, we obtain the PL intensities according to

$$I_{x(y)} \propto \sum_i e^{-\frac{E_i}{k_B T}} (S_{L,e} (\lambda_b^i + (-) \lambda_a^i) + S_{R,e} (\lambda_d^i + (-) \lambda_c^i))^2 \delta(E_i - \hbar\omega), \quad (9)$$

where  $\lambda_\alpha^i$  is the coefficient of the configuration  $|\alpha\rangle$  in the superposition of the eigenstate  $|i\rangle$  after diagonalizing the Hamiltonian,  $k_B$  is the Boltzmann constant,  $\omega$  is the frequency of the emitted photon, and  $S_{n,j}$  is the overlap between the electron and hole wave functions of states  $|n\rangle$  and  $|j\rangle$ , respectively ( $S_{n,j} = \int d^3\mathbf{r} \Phi_n^{h*}(\mathbf{r}) \Phi_j^e(\mathbf{r})$ ).

Figure 3 shows the PL spectra versus varied bias fields for the same case as Fig. 2, at a temperature of 10 K. For the reason of visibility, we use a Gaussian function with large standard deviation  $\gamma = 18$   $\mu\text{eV}$  to model the width broadening of the spectral lines [10]. The normalized intensities of the polarized PL lines and the corresponding FSS's as functions of bias field are shown in Fig. 2(b). There, significant reduction of the splitting, down to 50% of the magnitude of such energy in single dot cases, is observed as the double-dot system is electrically switched from the regime of off-resonance ( $eFz_0/V_{eh} \gg 1$  or  $\ll 1$ ) to that of near-resonance ( $eFz_0/V_{eh} \sim 1$ ) regime. As suggested in Ref. [12], such a controllability of the optical FSS's would be a key element in the realization of dot-based entangled photon pair emitters. We also find that in Fig. 2(b) the intensities of the emission lines basically follow the similar dependence of applied bias as that of FSS. This in-



**Figure 3** (online colour at: www.pss-b.com) Polarized photoluminescence (PL) spectra from single excitons in double quantum dots with an interdot distance of  $z_0 = 10$  nm under bias field control. Blue dashed lines indicate the PL spectra of  $\pi_x$  linearized polarization and the red solid lines indicate that of  $\pi_y$  polarization.

dicates that, as a FFS is tuned to be smaller, the oscillator strength of the e–h recombination becomes weaker and the intrinsic broadening of emission lines becomes also smaller. It turns out that the upper limit of FSS required for generating entangled photon-pairs is decreased. Nevertheless, the intensity of the polarized PL lines presented here might be underestimated because the penetration of electron wave function into the right dot is disregarded within the 3-orbital model.

**5 Summary** In summary, we present a theory of e–h EI in vertically coupled DQDs and explore the possibility of controlling the FSS of X states by utilizing the tuning parameters such as inter-dot distance and applied bias field. The developed theory was built up from the full interacting Hamiltonian of DQDs, taking into account both of the macroscopic and microscopic natures of particle wave functions. Compact formulations of e–h EI terms as functions of the material properties, geometry structure of DQDs and the strength of applied bias field are explicitly derived. The feasibility of the FSS control is supported by the numerically calculated energy and polarized PL spectra of spin Xs in DQDs. Under some determinable conditions, the controlled energy splitting could almost reach the upper limit of intrinsic radiative linewidth required by the application of polarization-entangled photon pairs source, as reported by Hafenbrak and Hudson et al. in Refs. [11] and [12].

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