Mechanically encoded single photon sources: 

Stress-controlled excitonic fine structures of droplet epitaxial quantum dots

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Abstract

We theoretically investigate the fine structure and photonic properties of valence-band-mixed (VBM) excitons in mechanically controlled GaAs/AlGaAs droplet epitaxial quantum dots (QDs). Supported by fully numerical calculations based on the multi-band $k \cdot p$ theory, a effective theory of fine structures of QDs is established in terms of the pseudo-spin of exciton coupled to the generalized effective fields yielded by geometry elongation and uniaxial stress. Based on the formalisms, the free access of any bright exciton qubit states is predicted to be possible by the appropriate design of the stressing and the subsequent polarized optical excitation of the QD. The degrees of entanglement for the emitted photon pairs from individual stress-controlled QDs are shown always non-maximal, with the upper bound set by the intrinsic VBM.

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Excitonic fine structures (FS’s) of semiconductor quantum dots (QDs) have been realized as an essential feature of advanced QD-based photonic applications, such as entangled photon pair emitters [1–5] and exciton-qubit gates. [6, 7] The realization of entangled photon pair emitters based on QDs has been for a long time a challenging task because it needs to retain the degeneracy of exciton doublet states, which is however likely lift by any slight symmetry breakings of QD structure. Remarkably, Trotta et al. have recently demonstrated an efficient way to recover, universally, the degeneracies of exciton doublet states of asymmetric QDs by means of electrical and mechanical controls. [4] The success in the exploitation of mechanical stress control paves a way to extend the usefulness of QD photon emitters with the potential integrations with micro electro-mechanical systems (MEMS) and nano-acoustics. [8] Besides, increasing attention is being paid to the exploitation of the exciton doublets of QDs as spin qubits that are convertible to flying photonic qubits for use in optical communications. [6, 7] Unlike an entangled photon pair emitter, an efficient control of exciton qubit yet needs an energy level anti-crossing in the fine structure of a QD where the exciton superposition states are highly tunable. [10]

With the great success in the fabrication of single QD photon sources, the next generation technology would be aiming at the realization of scaled-up photon generation architecture, [11] which will certainly result in a need for simple but still quantitatively useful models of QD photon sources. Sophisticate theories such as empirical pseudo-potential methods or atomistic tight binding models have been employed to calculate the fine structures of individual QDs, which however rely on large scaled numerical computations and are hardly extended for scaled up photonic systems. [12–15]

This Letter presents an effective model of excitonic fine structures of droplet epitaxial GaAs/AlGaAs QDs under uni-axial stress control, based on the Luttinger-Kuhn four-band $k \cdot p$ theory, which is quantitatively validated by fully numerical results and concise so as to be useful for scaled-up photonic architectures. The key advance made in the theory is the appropriate incorporation of the stress-enhanced VBM of exciton into the e-h exchange interactions (both long- and short-ranged parts) for highly stressed elongated QDs. The theory predicts that the entangled photon pairs emitted from stress-controlled are always non-maximally entangled (referred to as hyper-entanglement) and could be useful for loophole-free tests of Bell inequality. In addition, mechanical preparation of exciton fine structure states prior to a polarized photo-excitation is suggested to be a feasible way to
realize mechanically encoded photon sources.

We begin with the generalized Hamiltonian for an interacting exciton in a QD, \[ H_X = \sum_{i_e} E_{i_e} c_{i_e}^+ c_{i_e} + \sum_{i_h} E_{i_h} h_{i_h}^+ h_{i_h} - \sum_{i_e,i_h,k,l} V_{i_e,j_h,k,l} c_{i_e,j_h,k,l}^+ h_{i_h,k,l} c_{i_e} + \sum_{i_e,j_h,k,l} V_{i_e,j_h,k,l}^{e,h} c_{i_e,j_h,k,l}^+ h_{i_h,k,l} c_{i_e}, \] where \( c_{i_e} \) and \( c_{i_h} \) (\( h_{i_h}^+ \) and \( h_{i_h} \)) are the particle creation and annihilation operators for the electron orbital \( i_e \) (the hole orbital \( i_h \)), \( V_{i_e,j_h,k,l}^{e,h} \) is the matrix elements of the exciton Hamiltonian. The intensity \( I_n(\vec{e}) \) of the e-h exchange interactions \( (e-h \) exchange interactions), \( E_{i_e}^{e,h}(\psi_{i_e}^e,\psi_{i_h}^h) \) denote the eigen energy (wave function) of a single electron and single hole in the QD, respectively. Within the single band model (four-band \( k \cdot p \) model), the single-electron (hole) wave function is written as \( \psi_{i_e}^e = g_{i_e}^e u_{s_z}^e \) with \( s_z = \pm \frac{1}{2} \) \( (\psi_{i_h}^h = \sum_{j_z=\pm\frac{1}{2}} g_{i_h,j_z}^h u_{j_z}^h) \) which comprise the slowly varying envelope wave functions \( g_{s_z}^{e,h} \) and microscopic Bloch functions \( u_{s_z}^{e,h} \). Following Ref.\[17\], we consider GaAs/AlGaAs DE-QDs shaped by a Gaussian profile \( \exp(-\frac{x^2}{\Lambda_x^2} - \frac{y^2}{\Lambda_y^2}) \), to which is applied a uniaxial stress of magnitude \( \sigma \) in the direction of \( \vec{n}_\sigma = (\cos \phi_\sigma, \sin \phi_\sigma, 0) \), as depicted in Fig. 2. Numerically, the energy spectrum and the envelope functions for an electron (a hole) \( g_{s_z}^e (\{g_{s_z}^h\}) \) are obtained by solving the single-band (four-band) Schrödinger equation using the finite difference method. (See Ref.\[18\] for technical details and used parameters.)

To calculate the FS of an exciton in a QD, we first take the lowest valence-band-mixed BX configurations as the basis, \( |\downarrow_e \rangle |\uparrow_h \rangle \) and \( |\uparrow_e \rangle |\downarrow_h \rangle \), and then construct the corresponding \( 2 \times 2 \) matrix of the interacting exciton Hamiltonian.\[18\] Here, for brevity, spin arrows \( \uparrow, \downarrow \) \( (\uparrow_h, \downarrow_h) \) are used to label the lowest electron (VBM hole) states. The e-h exchange interactions yield the non-vanishing off-diagonal elements of the matrix, \( V_{i_e,j_h,k,l}^{e,h} \), coupling the two BX configurations and causing the FSS of the BX states, \( |S| = 2 |V_{i_e,j_h,k,l}^{e,h} \rangle \langle i_e,j_h,k,l |. \) Numerically, the matrix elements \( V_{i_e,j_h,k,l}^{e,h} \) are divided into the short- and long-ranged parts according to the averaged Wigner-Seitz radius,\[15\] and computed following the approaches used in Refs.\[20,21\] (See \[18\] for detailed theory). The energy spectrum \( \{E_{n}^X\} \) and the eigen-states \( \{\Psi_n^X\} \) of a VBM exciton in a stressed QD are then obtained by diagonalizing the matrix of the exciton Hamiltonian. The intensity \( I_n(\vec{e}) \) of the e-polarized light emission from the \( n \)th exciton state is calculated by using Fermi’s golden rule, \( I_n(\vec{e}; \omega) \propto |\langle 0 | P_{\vec{e}}^n | \Psi_n^X \rangle|^2 \delta (E_n^X - \hbar \omega) \), where \( |0\rangle \) is the vacuum state, \( \vec{e} = (\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta) \) is the unit vector of polarization, and the polarization operator is defined as \( P_{\vec{e}}^n = \sum_{ij} D_{ij}^e c_i \hbar_j \) in terms of the.
dipole matrix elements $D_{ij}^e = \langle \psi^h_i | \hat{e} \cdot \vec{r} | \psi^e_j \rangle = \sum_j \langle g^h_{i,j,z} | g^e_{j,s,z} \rangle \langle u^h_{j,z} | \hat{e} \cdot \vec{p} | u^e_{s,z} \rangle$. The explicit formalisms for $\langle u^h_{j,z} | \hat{e} \cdot \vec{p} | u^e_{s,z} \rangle$ can be found in Ref. [22].

For general description, the polarization axis close to the $x$-axis ($y$-axis) is specified as $\hat{n}_\alpha$ ($\hat{n}_\beta$), as depicted in Fig. 1(b), and the corresponding exciton state as $|\Psi^X_\alpha\rangle$ ($|\Psi^X_\beta\rangle$) for a stressed QD. To quantify the optical polarization anisotropy of an excitonic FS, the degree of linear polarization (DOP) is defined as $DOP \equiv I_\alpha - I_\beta \over I_\alpha + I_\beta$, where $I_\alpha \equiv I_\alpha(\hat{e} = \hat{n}_\alpha)$, and the energy difference between the BX doublet, $S \equiv E^X_\alpha - E^X_\beta$. Notably, the signs of $S$ and DOP can be either positive or negative, reflecting the energy order and the intensity ratio of the $\hat{n}_\alpha$- and $\hat{n}_\beta$- polarized emission lines.

Figures 2 (a),(c), and (d) show the numerically calculated polarized emission spectra, the FSS’s and the DOP’s of an $x$-elongated GaAs/AlGaAs QD of $H = 9\text{nm}$, $\Lambda_x = 14\text{nm}$ and $\Lambda_y = 12.7\text{nm}$ under uniaxial stresses aligned along the elongation axis ($\phi_\sigma = 0^\circ$). In the absence of any stress, the low-energy (LE) emission line of the QD is $x$-polarized and slightly brighter than the high-energy (HE) line (see the middle plots in Fig. 2 (a)), characterized by $S \sim -6.3\mu\text{eV} < 0$ and DOP $\sim +2.5\% > 0$ (See Figs. 2 (c) and (d)). Applying a compressive uniaxial stress ($\sigma < 0$ and $\phi_\sigma = 0^\circ$) makes the $x$-polarized line even brighter and blue-shifted. It turns out that the DOP becomes greater and the $S$ changes from negative to positive as $\sigma < -0.1\text{GPa}$ (where $S > 0$, DOP $> 0$). By contrast, the application of a tensile stress ($\sigma > 0$ and $\phi_\sigma = 0^\circ$) makes the $x$-polarized line darker and energetically even lower, leading to $S < 0$ and DOP $< 0$. Disabling the VBM (by setting $S = R = 0$ in the $k \cdot p$ model) causes the calculated DOP to vanish and reverses the order of energies of polarized lines (Fig. 2 (b) and the dashed line in (c)). The crucial role of VBM in the FSs of a stressed QD is further elucidated by extracting the optically active portion of the light-hole (LH) component, $\beta_{HL} \equiv |\langle g^e_{s,=\pm1/2} | g^h_{j,s,=\pm1/2} \rangle|^2$, from the exciton wave function, and noting that it exhibits nearly the same stress-dependence as those of $S$ and DOP (Figs. 2 (c) and (d)).

If the applied stress is misaligned with the elongation axis ($\phi_\sigma \neq 0^\circ$), then the optical axes rotate away from the $x$- or $y$-axes,[23] and the magnitudes and signs of the $S$ and DOP, which are $S > 0$ and DOP $> 0$ in the both compressive- and tensile-stress regimes, change, as shown in Figs. 2 (e)-(h) for $\phi_\sigma = 30^\circ$.

As a first step in the analysis, we follow the approach used in Ref.[24], which takes the lowest $e$- and $h$- harmonic oscillation (HO) wave functions ($\phi^X_{000} \equiv \phi^X_S =$
\[ \chi = e/HH/LH \] as the particle envelope functions. Then, the employed VBM BX configurations are expanded as \( c_{\uparrow}^{e} \phi_{S}^{e} |0\rangle \equiv | \downarrow_{e} \rangle | \uparrow_{e}^{\prime} \rangle \approx | \phi_{S}^{e} \rangle | \phi_{S}^{\prime} \rangle \) and \( c_{\uparrow\uparrow}^{e} \phi_{S}^{\prime} |0\rangle \equiv | \uparrow_{e} \rangle | \psi_{h} \rangle \approx | \phi_{S}^{\prime} \rangle | \phi_{h} \rangle \) - \( \tilde{\beta}_{HL} | \phi_{S}^{LH}; \uparrow_{h} \rangle \), where \( \tilde{\beta}_{HL} \equiv \langle \phi_{S}^{LH} | \hat{R} | \phi_{S}^{LH} \rangle \) and \( \Delta_{HL} \equiv \langle \phi_{S}^{LH} | \hat{P} - \hat{Q} + V_{QD} | \phi_{S}^{LH} \rangle - \langle \phi_{S}^{LH} | \hat{P} + \hat{Q} + V_{QD} | \phi_{S}^{LH} \rangle \).\[24\] The operators \( \hat{R} \), \( \hat{P} \), and \( \hat{Q} \) are defined in Ref.\[18\] following the standard Luttinger-Kohn \( k \cdot p \) theory. On the theoretical base, a generalized theory for the excitonic FS of stressed QDs is presented and used to analyze the numerical data.\[18\] For clarity, the formalisms for the QDs with \( l_{\alpha}^{0} = l_{\alpha}^{HH} = l_{\alpha}^{LH} \equiv l_{\alpha} \) are shown below.

In the single HO-orbital approximation, the off-diagonal matrix elements \( V_{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}}^{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}} = \tilde{\Delta}_{eff} \) can be formulated as

\[ \tilde{\Delta}_{eff} = -\Delta_{1} + \Delta_{VBM} \equiv \Delta_{eff} e^{-\theta_{eff}} , \]

as presented in Ref.\[24\]. Here, \( (-\Delta_{1}) = V_{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}}^{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}} \) is the attractive long ranged \( e-h \) exchange interaction between pure heavy-hole (HH) exciton configurations, and the correcting term,

\[ \tilde{\Delta}_{VBM} = \frac{2}{\sqrt{3}} \tilde{\beta}_{HL} E_{X}^{S}, \]

originates from the repulsive short-ranged interaction, \( E_{X}^{S} \equiv V_{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}}^{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}} = -\sqrt{3} V_{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}}^{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}} = V_{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}}^{\epsilon_{e}, \phi_{h}, \phi_{h}, \epsilon_{e}} \), which is namely the splitting of BX- and DX- levels,\[21, 24\] and appears in Eq.\[11\] via the VBM that is quantified by \( \tilde{\beta}_{HL} \) here.

In the parabolic model of QD, the VBM-term \( \tilde{\beta}_{HL} \) is explicitly expressed as

\[ \tilde{\beta}_{HL} = \left[ \frac{\sqrt{3}h^{2} \gamma_{3}}{4m_{0}} \frac{(1 - \eta^{-2})}{l_{y}^{2}} - \frac{|d| s_{44}}{4} \sigma \cos 2\phi_{\sigma} \right. \]

\[ + \left. i \frac{\sqrt{3} |b| (s_{11} - s_{12})}{2} \sigma \sin 2\phi_{\sigma} \right] / \Delta_{HL} , \]

where \( \Delta_{HL} \approx \frac{h^{2} \gamma_{2}}{m_{0}} \cdot \frac{1}{l_{y}^{2}} - |b| (s_{11} - s_{12}) \sigma \) is the energy difference between the HH- and LH-levels and \( \eta \equiv l_{z}/l_{y} \) is defined to parametrize the QD elongation, \( \Delta_{1} = \frac{1}{4\pi \sqrt{2} \eta} e^{\frac{3\gamma_{3} \eta}{4l_{y}}} \left( \frac{3\gamma_{3} \eta}{4l_{y}} \right)^{2} \text{erfc} \left( \frac{3\gamma_{3} \eta}{4l_{y}} \right) \),\[20\] and \( E_{X}^{S} \approx \frac{1}{2} \frac{a_{B}^{*} \cdot 3 \tilde{\Delta}_{xc \text{, bulk}}^{e}}{\sqrt{8\pi l_{z} l_{y} l_{x}}} . \) Here, \( \gamma_{1} = 7.1, \gamma_{2} = 2.02, \gamma_{3} = 2.91, \) \( b = -1.7eV, \) and \( d = -4.55eV \) are the \( k \cdot p \) parameters, \( s_{11} = 0.0082 \text{GPa}^{-1}, s_{12} = -0.002 \text{GPa}^{-1} \), and \( s_{44} = 0.0168 \text{GPa}^{-1} \) the elastic compliance constants, \( a_{B}^{*} = 11 \text{nm} (\Delta_{xc \text{, bulk}}^{e} = 20 \text{eV}) \) is the effective Bohr radius (the BX- and DX-level splitting) of exciton in bulk GaAs, \( E_{P} = 28.8eV, \) and \( E_{g} = 1.519eV \) is the energy gap of GaAs.\[18, 23, 26\]
Taking the eigen states of the un-stressed QD, \(|\Psi^X_x\rangle = \frac{1}{\sqrt{2}} (|\downarrow_x\rangle |\uparrow^0_y\rangle + |\uparrow_x\rangle |\downarrow^0_y\rangle)\) and \(|\Psi^X_y\rangle = \frac{i}{\sqrt{2}} (|\downarrow_y\rangle |\uparrow^0_x\rangle - |\uparrow_y\rangle |\downarrow^0_x\rangle)\) which are optically are x- and y-polarized, respectively, as known by Ref.\([24]\) as basis, the exciton Hamiltonian for a QD with an arbitrary stress can be reformulated as

\[ H'_X = \vec{\sigma} \cdot \vec{\Omega}_{\text{eff}} \]

where, for brevity, the effective Hamiltonian is offset by the averaged energy of the BX doublet, \(H'_X = H_X - E^{(0)}_X\), and \(\vec{\sigma} = (\sigma_1, \sigma_2, \sigma_3)\) is the Pauli matrix vector for exciton spin, which is shown coupled to the effective field

\[ \vec{\Omega}_{\text{eff}} \equiv (\Omega_1, \Omega_2, \Omega_3) = (-3\bar{\Delta}_{\text{eff}}^{\text{ex}}, 0, 0, \text{Re} \Delta_{\text{eff}}^{\text{ex}}), \]

with the components, \(\Omega_1\) and \(\Omega_3\), of the stress-resulting effective e – h exchange interaction. In the formalism, the exciton eigen states \(|\Psi^X_x\rangle\) with energies \(E^X_+ = E^X_0 \pm |\vec{\Omega}_{\text{eff}}| = E^{(0)}_X \pm \Delta_{\text{eff}}\) can be represented by Bloch vectors, \(|\Psi^X_x\rangle = (\cos \frac{\theta_X}{2}, e^{-i\phi_x} \sin \frac{\theta_X}{2})\) and \(|\Psi^X_y\rangle = (\sin \frac{\theta_X}{2}, -e^{-i\phi_x} \cos \frac{\theta_X}{2})\), respectively, mapped onto the Bloch sphere with geometrical angles \(\theta_X = \theta_{\text{eff}}\) \((\theta_X = 360^\circ - \theta_{\text{eff}})\) and \(\phi_X = 0^\circ\) \(\phi_X = 180^\circ\) as \(\theta_{\text{eff}} = \{0^\circ, 180^\circ\}\) (as \(\theta_{\text{eff}} = \{180^\circ, 360^\circ\}\)). Equations (4) and (5) provide a base of an exciton Bloch equation for general dynamic studies, and show that the spin orientation of an exciton can be controlled by appropriately designing \(\vec{\Omega}_{\text{eff}}\) by magnetic, geometric, or even mechanical means.

In the next stage of the analysis, the model is further improved by including higher HO orbitals relevant to the stress-dependent exciton wave functions, leading to the rescaled long ranged interaction, \(\Delta_1 \rightarrow \Delta'_1\), as detailed by Ref.\([18]\). The results obtained by the improved model are quantitatively even more consistent with the fully numerical one, as one can see in Fig.2 (c),(d), (g) and (h). In the model calculations, we take the parameters for the considered QD, \(l^c_x = 5.99\) nm, \(l^c_y = 5.66\) nm, \(l^e_x = 2.99\) nm, \(l^{HH}_x = 6.66\) nm, \(l^{HH}_y = 6.11\) nm, and \(l^{HH}_z = 2.33\) nm \((l^{LH}_{x,y} = r^{-1}_{\perp} l^{LH}_{x,y} \text{ and } l^{LH}_z = r^{-1}_{\perp} l^{LH}_z)\) are inferred with \(r_{\perp} = (\frac{21+2\gamma_2}{21-2\gamma_2})^{1/4}\) and \(r_{\parallel} = (\frac{21+2\gamma_2}{21-2\gamma_2})^{1/4}\), which are extracted by fitting the numerically calculated wave functions. The intensities of the \(\hat{e}\)-polarized \((\hat{e} = (\cos \phi, \sin \phi, 0))\) emitted lights from the BX doublet states are derived as \(I_+ \propto |A_+|^2 \cos^2(\phi - \phi_+)\) and \(I_- \propto |A_-|^2 \sin^2(\phi - \phi_+)\), where \(\phi_0 = \frac{\theta_X}{2} + \frac{\phi_0}{2} + \delta \phi \pm\) with \(\delta \phi \pm = \tan^{-1}\left(\frac{\mp \Delta_{\text{eff}} \cos \theta_{\text{eff}}}{\pm \Delta_{\text{eff}} \cos \theta_{\text{eff}} / \Delta_1 \cos \theta_{\text{eff}}}\right)\) and \(A_\pm = \left(1 \pm \frac{\Delta_{\text{eff}} + \Delta_1 \cos \theta_{\text{eff}}}{2E_N} \right)^2 + \left(\frac{\Delta_{\text{eff}} \sin \theta_{\text{eff}}}{2E_N}\right)^2\) \(1/2\).

Thus, the polarization of the HE (LE) exciton state \(|\Psi^X_+\rangle (|\Psi^X_-\rangle\) is shown along the optical axis with the angle around \(\sim \theta_{\text{eff}}/2\), slightly modified by \(\delta \phi_+ (\delta \phi_-)\), to the x-(y)-axis.

For a QD under an uniaxial stress with \(\phi_0 = 0^\circ\), \(I_x = I_0(1 + \beta_{HL}/\sqrt{3})\) and \(I_y = I_0(1 - \sqrt{3})\).
\[ \beta_{HL}/\sqrt{3} \] can be shown, resulting in the DOP= \( \frac{2}{\sqrt{3}} \beta_{HL} \), and \( S = 2(-\Delta_1 + \Delta_{VBH}) = 2(-\Delta_1 + \beta_{HL}E_X^S) \). Thus, a highly compressive (\( \sigma < 0 \)) stress that makes \( \beta_{HL} \propto -\sigma \cos 2\phi_\sigma > 0 \) significantly large and \( \Delta_{xc}^e \sim \Delta_{VBH} = \beta_{HL}E_X^S > 0 (\theta_{eff} = 0^\circ) \) leads to DOP > 0 and \( S > 0 \). Likewise, for a QD under significant tensile stress, \( \beta_{HL} < 0 \) and \( \Delta_{xc}^e \sim \beta_{HL}E_X^S < 0 (\theta_{eff} = 180^\circ) \) can be shown, resulting in DOP < 0 and \( S < 0 \). The reversal of the signs of \( S \) and DOP (which indicates the reversal of the order of the energy and brightness of the x- and y-polarized lines) in the highly compressive and tensile regimes is caused by the significant short-ranged interaction \( \Delta_{VBH} \) arising from VBM, as shown by comparing Figs.2(c) and (d). The analysis also elucidates the S-DOP correlated feature (via the common underlying VBM, \( \beta_{HL} \)) observed in Fig.3(a) from a series of elongated QDs with various stresses.

The formalism, \(|S| = 2\sqrt{\Re(\Delta_{xc}^{eH} - \Re(\Delta_{xc}^{eV}))}, \) shows that the FSS of a stressed QD never vanish as long as \( \Re(\Delta_{xc}^{eH}) < 0 (i.e. \phi_\sigma \neq 0^\circ, 90^\circ) \). Therefore, as a prerequisite for the generation of entangled photon pair, the vanishing of the FSS \((S = 0)\) of a QD is achievable only when the applied stress is exactly parallel or perpendicular to the axis of elongation and \( \Delta_{VBH} = \Delta_1 (\neq 0) \) such that \( \Re(\Delta_{xc}^{eH}) = 0 \). The latter condition indicates that \( \beta_{HL} \neq 0 \) and therefore the DOP \((\propto \Delta_{VBH}) \neq 0 \). Owing to the non-zero DOPs, the emitted photon pairs of stressed QDs with \( S = 0 \) are in non-maximally entangled two-photon states, given by \(|n = 2\rangle_{ph} = (|HH\rangle + \epsilon|VV\rangle)/\sqrt{1 + \epsilon^2}, \) with the degree of entanglement \( \epsilon = \frac{1-DOP}{1+DOP}. \) where \( H \) (V) indicates the x-(y-) polarized photon. Such a non-maximally entanglement \((\epsilon \neq 1)\) has been shown to be advantageous for reducing of the required detector efficiencies for loophole-free tests of Bell inequalities.\[27, 28\] Figure 3(c) plots the degree of entanglement \( \epsilon \) as a function of the elongation \( \eta \) of the stressed QDs, which can be as low as \( \epsilon \sim 0.6 \) for \( \eta = 1.2 \).

Rotating the axis of stress from that of QD elongation \((\phi_\sigma \neq 0)\) adds an additional phase angle \((\theta_{eff} \neq 0)\) to the effective interaction \( \tilde{\Delta}_{xc}^{eH} \equiv \Delta_{xc}^{eH}e^{-i\theta_{eff}} = -\Delta_1 + \frac{2}{\sqrt{3}}(\beta_{HL,k} + \beta_{HL,\epsilon})E_X^S = -\Delta_1 + \Delta_{VBH,k} + \Re(\Delta_{VBH,\epsilon}) + i\Im(\Delta_{VBH,\epsilon}) \) since \( \Re(\Delta_{VBH,\epsilon}) \propto (-\sigma \cos 2\phi_\sigma) \) and \( \Im(\Delta_{VBH,\epsilon}) \propto (\sigma \sin 2\phi_\sigma) \). If the rotation of the stress axis is significant, such that \( \cos 2\phi_\sigma < 1 \), then \( \cos \theta_{eff} \propto (-\Delta_1 + \Delta_{VBH,k}) \approx -\Delta_1 < 0 \) and \( \sin \theta_{eff} \propto -\Im(\Delta_{VBH,\epsilon}) \approx (-\sigma \sin 2\phi_\sigma) \) result. Accordingly, one can infer that \( \theta_{eff} = \{90^\circ, 180^\circ\} \) as \( \sigma < 0 \) and \( \theta_{eff} = \{180^\circ, 270^\circ\} \) as \( \sigma > 0 \). Thus, under a rotated highly compressive (tensile) stress \( \sigma < 0 \) \((\sigma > 0)\), the HE emission line of \( I_+ = I_\beta \) of a QD brighter and polarized along the optical axis with \( \phi_+ \approx \theta_{eff}/2 = \{45^\circ, 90^\circ\} \) \((\phi_+ \approx \theta_{eff}/2 = \{90^\circ, 135^\circ\})\), as shown in the inset of Fig. 4(a) for the
stressed QD with \( \phi_\sigma = 45^\circ \). Recall that 
\[
I_\pm \propto |A_\pm|^2 = \left[ (1 \pm \frac{\Delta_{eff} + \Delta_1 \cos \theta_{eff}}{2E_X})^2 + \left( \frac{\Delta_1 \sin \theta_{eff}}{2E_X} \right)^2 \right],
\]
which clearly shows that, if the applied stress is sufficiently high such that \( \Delta_{eff} \sim \Delta_{VBM} \gg \Delta_1 \), the HE emission line is certainly brighter than the LE one, and \( I_+ > I_- \). In summary of the above analysis, \( I_\beta = I_+ > I_\alpha = I_- \) and, therefore, \( S < 0 \) and DOP < 0 for the QD highly stressed with \( \phi_\sigma = 30^\circ \) in Fig. 2 (e)-(h). In short, with a significant rotated stress, the HE radiative line of a stressed QD is always brighter and polarized near the \( y \)-axis, which lies in either the quadrant I or quadrant II of the polar plot, depending on the type of stress (tensile or compressive).

From our derived formalism for \( \phi_\pm \), as \( \theta_{eff} \neq 0 \) (due to \( \phi_\sigma \neq 0 \)), the optical axes of the two emission lines are not really perpendicular to each other \( (\delta \phi_+ \neq \delta \phi_-) \), as recently experimentally observed.\[29\] Figure 4 (a) shows the angle of the \( \hat{n}_\alpha-(\hat{n}_\beta-) \) optical axis, \( \phi_+ \) (\( \phi_- \)), from the \( x \)-(\( y \)-) axis and the corresponding phase angle of effective \( e-h \) exchange interaction, \( \theta_{eff}/2 \), of the uniaxially stressed QD with various \( \sigma \) but fixed \( \phi_\sigma = 45^\circ \). The slight difference between \( \phi_\pm \) indicates the non-orthogonality of the optical axes.

Experimentally, an exciton superposition state in the FS of a QD has been demonstrated to be initially created by a quasi-resonant laser pulse with appropriate polarization, and then evolves (within the coherence time) in a free precession, which can be, geometrically, described by a circle on the Bloch sphere about the axis connecting the exciton eigen states. Applying an external stress to a QD rotates the exciton eigen states at the north and south poles of the Bloch sphere towards the equator by an angle \( \theta_X \), as exemplified by the schematics in Fig. 4(b), and tilt the circle on which the superposition exciton state evolves. Figure 4 (b) shows the phase angles \( \theta_X \) of the exciton states of the stressed QD with various \( \sigma \) and \( \phi_\sigma \), which are shown highly stress-tunable. The dynamics of such mechanically encoded exciton states can be monitored by optically measuring the polarization projection using the techniques presented in Ref.\[6, 7\]. Figure 4(c) shows the \( x \)-polarization (\( H- \)) projection \( P^H_R(t) \) of the exciton superposition state of the QD stressed with fixed \( \phi = 45^\circ \) that is optically initialized by a right-handed circular (\( R- \)) polarized laser \( (|\Psi^X_R(t = 0)\rangle = C^R_+ |\Psi^X_+ \rangle - iC^R_- |\Psi^X_- \rangle \) where \( C^R_\pm = (\langle 0 |P^R_\pm |\Psi^X_\pm \rangle)^* \) (ideal coherence is assumed). Analytically, 
\[
P^H_R(t) \equiv |\langle 0 |P^H_R |\Psi^X_R(t) \rangle|^2 = \frac{1}{2}[(1 + DOP)^2 \cos^2 \phi_+ + (1 - DOP)^2 \sin^2 \phi_- - 2(1 - DOP^2) \cos \alpha \sin \delta \sin(\frac{\alpha}{2} - \phi_+ + \phi_-)]
\]
can be shown to be a function of the stress-dependent DOP and \( S \), where \( \phi_+ = \phi_\beta \) (\( \phi_- = \phi_\alpha \)) is the angle of the optical axis for \( |\Psi^X_+ \rangle = \Psi^X_\beta \) \( (|\Psi^X_- \rangle = \Psi^X_\alpha) \). Clearly, with varying the stress strength, the same initialized
exciton state of the stressed QD evolve in distinctive ways. The inset in Figure 4(c) depicts the path of the evolving exciton state of the QD that is stressed with $\phi = 45^\circ$ and $\sigma = -0.2$ GPa.

This Letter demonstrates an effective theory for the fine structure states of bright excitons in droplet epitaxial QDs under mechanical control, which is quantitatively validated by fully numerical computations based on Luttinger-Kuhn four band $k \cdot p$ theory and extendible for scaled up photonic architectures. From the presented formalisms, the entanglement of the emitted photon pairs from individual QDs is predicted to be always non-maximal, with the upper bound of the degree of entanglement set by intrinsic valence band mixing. Prior mechanical control of the exciton fine structure states are shown to be feasible for realizing mechanically encoded photon sources.

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FIG. 1: (a) Schematics of a quantum dot (QD) photon source mounted on a piezoelectric actuator (PMN-PT) under a controlled uniaxial stress $\sigma$. (b) Polarized fine structures of exciton of a stressed QD without and with valence band mixing (VBM). (c) Polar plot of optical polarization and (d) polarized emission spectrum of a VBM exciton in a uniaxially stressed QD.
FIG. 2: (a) Calculated polarized emission spectra and the corresponding polar plots of of a $x$-elongated QD under uni-axial stresses along the $x$-axis ($\phi_\sigma = 0^\circ$) with strength $\sigma = 0, \pm 0.15\text{GPa}$. (b): as (a) but without the consideration of VBM. (c) Calculated $S(\equiv E_{\alpha=x} - E_{\beta=y})$ for the FS of the stressed QD as a function of $\sigma$. Red circles: fully numerical results obtained by $k \cdot p$ theory. Blue solid lines: results based on the model developed herein. Grey dashed line plots the $S$ without VBM. Dotted green line shows $\Delta_{VBM}$, which substantially affects the $S$ of the stressed QD in the presence of stress. (d) Degree of polarization DOP versus $\sigma$ of emission lines from the stressed QD. (e)-(h): as (a)-(b), but for $\phi_\sigma = 30^\circ$. 
FIG. 3: (a) $S$ versus DOP of fine structure emission lines from single excitons in elongated QDs of $\eta = 1, 1.05, 1.1, 1.2$ under uniaxial stress $\sigma = 0.1, 0.05, ..., -0.3$ GPa along the elongation axis of QD. The areas of the empty (filled) symbols reflect the magnitudes of the applied tensile (compressive) stresses. Note that the resulting DOPs of the stressed QDs with $S = 0$ are non-zero, leading to the non-maximal entanglement of the emitted photon pairs ($\epsilon < 1$). (b) Degree of entanglement $\epsilon$ of emitted photon pairs from the elongated QDs with stress-controlled vanishing $S$ as a function of the QD elongation $\eta \equiv l_x/l_y$. 

\[ H_{\text{HH}} = \frac{1}{\sqrt{1 + \epsilon}} \left(HH + \epsilon (VV)\right) \]
FIG. 4: (a) Optical polarization angles $\phi_\pm$ of the excitonic fine structure states of a stressed with $\phi_\sigma = 45^\circ$ as a function of the stress strength $\sigma$, which follow nearly the same $\sigma$-dependence as that of $\theta_{\text{eff}}/2$ (See Eq. (1) for the definition of $\theta_{\text{eff}}$). (b) Phase angles $\theta_X$ and $\phi_X$ of the exciton states of the stressed QD represented as Bloch vectors against the $\sigma$ and $\phi_\sigma$ of applied stresses, which are shown highly stress-tunable. (c) Time dependent $H$-polarization projection $P_H^R$ of the superposition exciton state of a $R$-polarized photo-excited QD that was mechanically prepared with uni-axial stresses of fixed $\phi_\sigma = 45^\circ$ but various $\sigma$. Note that the same initialized exciton states dynamically evolve in distinctive ways as $\sigma$ is changed. The inset depicts the path on the Bloch sphere that describes the dynamics of the exciton superposition state of the stressed QD with $\sigma = -0.2\text{GPa}$.