Semiconductor quantum dots (QDs) have been demonstrated as one of candidates for the entangled two-photon sources, which make them very attractive for applications in the fields of quantum teleportation and quantum computation.\footnote{Reference}\footnote{Reference}\footnote{Reference} Much efforts, e.g., thermal annealing and external field tuning\footnote{Reference} have been devoted to the reduction of the fine-structure splitting (FSS) of the intermediate exciton states, since a necessary condition in the proposal for a QD-based source of polarization entangled photon pairs is that the intermediate exciton states for the biexciton radiative decay are energetically degenerate.\footnote{Reference} FSS originated from the anisotropic electron-hole exchange interaction is directly determined by the anisotropic shape of the exciton envelope function. Obviously, exciton envelope function may be strongly influenced by the ionized shallow impurities in QDs, and consequently FSS might be changed. In this letter, we study the ground-state FSS of ionized hydrogenetic donor impurity-exciton complex in quantum dots and find that it could be largely reduced by the ionized hydrogenetic impurities as well as the external electric field.

Recently, with the advancement of QDs growth and measurement technics, it is possible to optically probe fine-structures of single magnetic impurity-doped semiconductor QDs.\footnote{Reference}\footnote{Reference} In this letter, the ground-state energy and FSS of ionized hydrogenetic donor impurity-exciton complex in semiconductor QDs under an external in-plane electric field are investigated, since shallow donor impurity is common and well studied in III-V semiconductors, e.g., Si-doped GaAs. The light-hole and spin-orbit-split $J = 1/2$ valence bands could be reasonably neglected in the calculations, since the heavy-hole component is dominant in the hole ground state of flat InGaAs QDs\footnote{Reference} and we mainly focus on the exciton ground states. Thus the exciton state is composed of 4 combinations of the valence band and the conduction band, i.e., \[|X\rangle = \sum_{m,s} \sum r_{e}, r_{h} \psi_{ms}(r_{e}, r_{h}) a_{m,r_{e}}^{j} a_{s,r_{h}}^{j} |0\rangle \] where the Wannier function representation of the creation and annihilation operators is used, \[\psi_{ms}(r_{e}, r_{h})\] is the exciton envelope function, and $m$ and $s$ are the $z$-component of the angular momentum of the heavy-hole valence band and the conduction band, respectively. The eigenvalue equation for $\psi_{ms}$ is given as

\[ \sum_{m',s',r_{e}', r_{h}'} [H_{1} + V_{ex}(e_{s}, m', s', r_{e}', r_{h}')] \psi_{ms}(r_{e}', r_{h}') = E \psi_{ms}(r_{e}, r_{h}), \]

with

\[ H_{1} = \delta_{e,s} \delta_{h,s'} \delta_{m,m'} [H_{e} + H_{h}] + e F \cdot (r_{e} - r_{h}) - \frac{e^{2}}{r_{e} - r_{h}} + V_{int}(r_{e}, r_{h}, q_{j}), \]

where $H_{k} = p_{k}^{2}/2m_{k} + U_{k}(r_{k})$ ($k = e, h$) is the single particle Hamiltonian, $U_{e}(U_{h})$ is the confinement potential for the hole (electron), $F$ is an external in-plane electric field, and $V_{int}(r_{e}, r_{h}, q_{j}) = \sum_{j=1}^{N} (e^{2}/|r_{e} - q_{j}| - e^{2}/|r_{h} - q_{j}|)$ is the Coulomb interaction between charge carriers and ionized impurity centers, $q_{j} = (x_{j}, y_{j})$ is the position of the $j$th ionized donor impurity, $N$ is the total number of ionized donor impurities. Similar to the assumption in Ref.\footnote{Reference} an in-plane anisotropic potential is used in modeling single QDs, i.e., $U_{c}(h) = U_{c} |\theta(h)/(2 - |y_{c}(h)|)/(a/2 - |x_{c}(h)|))$, where $a$ and $b$ are the lateral sizes of QDs, and $y_{c}(h)$ is the conduction (heavy-hole) valence band offset. Whether the geometric shape of single QDs is rectangular or elliptic will not change the qualitative results of this letter. $V_{ex}$ is the electron-hole exchange interaction.\footnote{Reference} The material parameters used in the calculations refer to Ref.\footnote{Reference} The computational procedure is that eigenfunctions of spin-independent $H_{1}$ is firstly calculated using diagonalization method with more than 4000 basis sets, and then FSS is obtained by calculating the matrix elements of $V_{ex}$ in the basis of eigenfunctions of $H_{1}$.

For simplicity, single ionized donor impurity is located along the $x$-axis, and the ionized impurity-exciton complex binding energy $E_{b}$ is defined as

\[ E_{b} = E(X) - E(D^{+}, X), \]

where $E(X)$ is the exciton ground state energy in QDs and $E(D^{+}, X)$ is the ground state energy of the ionized donor impurity-exciton complex in the same QD.\footnote{Reference} In
increases from zero, however, the binding energy initially
oscillator strength of the exciton transition is also shown.

At about
reduction of FSS is largely enhanced for the off-center
at the QD center, FSS is slightly reduced, relative to
results. For example, the ground-state FSS of two ionized donor
impurities-exciton complex in QDs, are shown
between electron-ionized donor and hole-ionized donor.
However, as the external field exceeds the effective local field, then oscillator strength is finally reduced as shown in
Fig. 1(a). When
becomes smaller and gradually approaches zero as
In Fig. 1(b), corresponding oscillator strength of the exciton transition is also shown.

including the exchange interaction
, the exciton
ground states are split into bright and dark doublets. FSS of bright doublet is mainly determined by the anisotropic shape of InGaAs QDs.\(^{12}\) FSS of QD2 with larger shape anisotropy and without impurity (66 \(\mu\)eV) is larger than that of QD1 with smaller shape anisotropy and without impurity (27 \(\mu\)eV), and calculated value of FSS is well consistent with recent experimental results.\(^{12}\) When there is an ionized donor impurity present at the QD center, FSS is slightly reduced, relative to the \(N = 0\) case, as shown in Fig. 1(c). Interestingly, reduction of FSS is largely enhanced for the off-center donor impurity. At about
FSS gets a minimal value, i.e., 17 \(\mu\)eV and 47 \(\mu\)eV for QD1 and QD2, respectively. Moreover, FSS could be further reduced when there are more than one ionized donor impurity. For example, the ground-state FSS of two ionized donor impurities-exciton complex in QD1 is only 6 \(\mu\)eV, with the two donor impurities position \(q_1 = (5.0\ \text{nm}, 1.0\ \text{nm})\) and \(q_2 = (5.0\ \text{nm}, -1.0\ \text{nm})\).

In Fig. 2(a), ground-state energies without the electron-hole exchange interaction for three different systems, i.e., (i) exciton without impurity, (ii) single ionized donor impurity-exciton complex, and (iii) two ionized donor impurities-exciton complex in QDs, are shown as functions of an external electric field along the x-axis. The single donor impurity position is (5.0 nm, 0 nm), and the two donor impurity positions are (5.0 nm, 1.0 nm) and (5.0 nm, −1.0 nm), respectively. The Stark shift of all three cases could be well approximated by a parametric model

\[ \Delta E(F) = \alpha F - \frac{1}{2} \beta F^2 + ... \]  

where \(\alpha\) and \(\beta\) are actually the exciton dipole and polarizability, respectively, along the external field.\(^{16}\) For the first case without ionized impurities, \(\alpha\) is zero and the Stark shift could be well fitted by the quadratic term. However, \(\alpha\) becomes nonzero for the second and third case because of the opposite Coulomb interactions between electron-ionized donor and hole-ionized donor. At small external electric field, the Stark shift could be well described by the linear and quadratic terms, while higher-order terms need to be taken into account at larger external electric field. Oscillator strength of the ground state shows interesting behaviors. For the first case, oscillator strength monotonically decreases as the external electric field, and its behavior is symmetric for the field in the positive and negative directions. For the last two cases, the behavior of the ground-state oscillator strength with the external field is clearly asymmetric. For \(F > 0\), external field partially counteracts the local field produced by the ionized impurities, and the overlap between electron and hole is initially enhanced. Therefore, the ground-state oscillator strength initially increases. However, as the external field exceeds the effective local field, then oscillator strength is finally reduced as shown in Fig. 2(b). For \(F < 0\), external field is in the same direction of the effective local field produced by the ionized impurities, and the oscillator strength monotonically decreases.

FSS of the three cases are shown as functions of the external electric field in Fig. 2(c). It can be seen that the behavior of exciton FSS in QDs with off-center ionized
impurities is greatly different from that of QDs without ionized impurities. For QDs without ionized impurities, FSS monotonically decreases with the external electric field. However, FSS of QDs with off-center ionized impurities shows asymmetric variations for external electric field in positive and negative directions. We note that FSS shows somewhat similar behaviors as functions of the external electric field with those of the oscillator strength, since both the oscillator strength and FSS are of the external electric field with those of the oscillator strength. Thus the results in this letter might be useful to explain the anomalous external electric-field dependence of FSS observed in the experiment.

In order to better understand the reduction of exciton FSS in QDs with ionized impurities, $\psi(r_e, r_h = r_e)$ of the ground state in QD1 along the $x$- and $y$-axis are shown in Fig. 3(a). For QD1 without ionized impurity and external field, the extension of $\psi(r_e, r_h = r_e)$ along the $x$-axis is slightly larger than that along the $y$-axis as clearly shown in Fig. 3(a). Thus the electron-hole long range exchange interaction is nonzero and FSS is calculated to be 27 $\mu$eV. When there are ionized donor impurities present as in the third case of Fig. 2 both the amplitude and shape of $\psi(r_e, r_h = r_e)$ are greatly changed due to the local electric field produced by the ionized donor impurities, and the extension along the $x$-axis becomes nearly identical to that along the $y$-axis as shown in Fig. 3(a). That is why FSS is largely reduced to only 6 $\mu$eV. In Figs. 3(b), 3(c), and 3(d), $\psi(r_e, r_h = r_e)$ of all the three cases in Fig. 2 are shown for several values of the external electric field, and it could be easily seen that both the amplitude and shape of $\psi(r_e, r_h = r_e)$ are largely changed by the external electric field as well as the ionized donor impurities, which clearly explains the anomalous behaviors of the external electric field and FSS in Figs. 2(b) and 2(c).
In summary, we study the ground-state FSS of ionized shallow donor impurities-exciton complex in anisotropic QDs, and find that it could be largely reduced by one or two off-center ionized donor impurities, which strongly influence the exciton envelope functions. Then anomalous Stark shifts and efficient tuning of FSS by the external electric field are clearly shown and discussed. The study will be helpful and interesting for the research on the QDs-based entangled-photon source.

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1. R.M. Stevenson, R.J. Young, P. Atkinson, K. Cooper, D.A. Ritchie, and A.J. Shields, Nature (London), 439, 179 (2006).
2. N. Akopian, N.H. Lindner, E. Poem, Y. Berlatzky, J. Avron, D. Gershoni, B.D. Gerardot, and P.M. Petroff, Phys. Rev. Lett. 96, 130501 (2006).
3. A.I. Tartakovskii, M.N. Makhonin, L.R. Sellers, J. Cahill, A.D. Andreev, D.M. Whittaker, J-P.R. Wells, A.M. Fox, D.J. Mowbray, M.S. Skolnick, K.M. Groom, M.J. Steer, H.Y. Liu, and M. Hopkinson, Phys. Rev. B 70, 193303 (2004); W. Langbein, P. Borri, U. Woggon, V. Stavarache, D. Reuter, and A.D. Wieck, Phys. Rev. B 69, 161301(R) (2004).
4. R.J. Young, R.M. Stevenson, A.J. Shields, P. Atkinson, K. Cooper, D.A. Ritchie, K.M. Groom, A.I. Tartakovskii, and M.S. Skolnick Phys. Rev. B 72, 113305 (2005); A. Greilich, M. Schwab, T. Berstermann, T. Auer, R. Oulton, D.R. Yakovlev, M. Bayer, V. Stavarache, D. Reuter, and A. Wieck, Phys. Rev. B 73, 045323 (2006).
5. R. Seguin, A. Schliwa, T.D. Germann, S. Rodt, K. Pötschke, A. Strittmatter, U.W. Pohl, and D. Bimberg, Appl. Phys. Lett. 89, 263109 (2006).
6. R.M. Stevenson, R.J. Young, P. See, D.G. Gevaux, K. Cooper, P. Atkinson, I. Farrer, D.A. Ritchie, and A.J. Shields, Phys. Rev. B 73, 033306 (2006).
7. K. Kowalik, O. Krebs, A. Lemaitre, S. Laurent, P. Senellart, P. Voisin, J.A. Gaj, Appl. Phys. Lett. 86, 041907 (2005).
8. B.D. Gerardot, S. Seidl, P.A. Dalgarno, R.J. Warburton, D. Granados, J.M. Garcia, K. Kowalik, O. Krebs, K. Karrai, A. Badolato, and P.M. Petroff, Appl. Phys. Lett. 90, 041101 (2007).
9. O. Benson, C. Santori, M. Pelton, and Y. Yamamoto, Phys. Rev. Lett. 84, 2513 (2000).
10. L. Besombes, Y. Léger, L. Maingault, D. Ferrand, and H. Mariette, Phys. Rev. Lett. 93, 207403 (2004); A. Kudelski, A. Lemaitre, A. Miard, P. Voisin, T.C.M. Graham, R.J. Warburton, and O. Krebs, Phys. Rev. Lett. 99, 247209 (2007).
11. W.D. Sheng, and P. Hawrylak, Phys. Rev. B 73, 125331 (2006).
12. E.L. Ivchenko, Phys. Status Solidi A 164, 487 (1997); T. Takagahara, Phys. Rev. B 62, 16840 (2000).
13. D. Xu, N. Zhao, and J.L. Zhu, J. Phys.: Condens. Matter 20, 045204 (2008).
14. B. Stébé, E. Assaid, F. Djurdjin, and S. Le Goff, Phys. Rev. B 54, 17785 (1996).
15. A.I. Tartakovskii, R.S. Kolodka, H.Y. Liu, M.A. Migliorato, M. Hopkinson, M.N. Makhonin, D.J. Mowbray, and M.S. Skolnick, Appl. Phys. Lett. 88, 131115 (2006); D.J.P. Ellis, R.M. Stevenson, R.J. Young, A.J. Shields, P. Atkinson, and D.A. Ritchie, Appl. Phys. Lett. 90, 011907 (2007).