**Statistical Properties of Exciton Fine Structure Splittings and Polarization Angles in Quantum Dot Ensembles**

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We propose an effective model to describe the statistical properties of exciton fine structure splitting (FSS) and polarization angle of quantum dot ensembles (QDEs). We derive the distributions of FSS and polarization angle for QDEs and show that their statistical features can be fully characterized using at most three independent measurable parameters. The effective model is confirmed using atomistic pseudopotential calculations as well as experimental measurements for several rather different QDEs. The model naturally addresses three fundamental questions that are frequently encountered in theories and experiments: (I) Why the probability of finding QDs with vanishing FSS is generally very small? (II) Why FSS and polarization angle differ dramatically from QD to QD? and (III) Is there any direct connection between FSS, optical polarization and the morphology of QDs? The answers to these fundamental questions yield a completely new physical picture for understanding optical properties of QDEs.

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**Introduction.** - The fine structure splitting (FSS) of excitons in self-assembled quantum dots (QDs) poses the major obstacle to the realization of entangled photon pairs from bichromophore cascade process, thus has been a subject of extensive investigation in the past decade \([1–3]\). Now it is quite clear that the FSS arises from the intrinsic nonequivalence along \([110]\) and \([110]\) directions in zinc-blende crystals, which reduce the symmetry of the underlying lattice from \(T_d\) to \(C_{2v}\) for pure circular lens-shaped QDs, and the other nonuniform effects such as local strain, shape irregularities, alloys and interface effects \([10, 11]\), which further reduce the symmetry to \(C_1\) for alloyed QDs \([12]\). A single external field, such as electric field \([13, 18]\), magnetic field \([2, 19]\), or anisotropic stress \([12, 20, 24]\), is insufficient to eliminate the FSS because the lower bound of FSS is generally much larger than the homogeneous broadening of the emission line (~1 μeV). To eliminate the FSS, two non-equivalent fields have to be combined \([8, 9]\). Generally, the FSSs depend strongly on the local details of QDs, thus is hard to be predicted in theories. For instance for two QDs with the same morphology but tiny difference in alloy atomistic arrangement, their FSS and polarization angle may be fairly different. Therefore we confront three fundamental issues, which are generally very challenging for understanding: (I) Why the probability of finding QDs with vanishing FSS is generally very small? (II) Why FSS and polarization angle differ dramatically from QD to QD? and (III) Is there any direct connection between FSS, polarization angle and morphology of QDs. These three questions are essential for understanding optical properties of QDEs.

This Letter is devoted to address these three fundamental issues. We propose an effective model to describe the statistical features of FSS and polarization angle and derive their corresponding distribution functions. We show that their statistical properties can be fully characterized using at most three independent measurable parameters. The effective model is then confirmed using atomistic pseudopotential calculations as well as experimental measurements for several rather different types of QDEs. Potential applications of the generic model is also discussed. These results yield a completely new physical picture for understanding optical properties of QDEs.

**Theoretical Model.** - An analytical model which can capture the true symmetry properties of QDs is required to better understand the statistical properties of QDEs. To this end, the recently developed phenomenological model in Ref. \([12]\) is well fitted to this problem. From the symmetry viewpoint, the Hamiltonian for a single QD can be written as \(H = H_{2v} + V_1\), where \(H_{2v}\) contains the kinetic energy and the potential with the crystal \(C_{2v}\) symmetry, and \(V_1\) is the perturbation potential with \(C_1\) symmetry. We define two eigenvectors of \(H_{2v}\) as \(|3\rangle = |\Gamma_2 + i\Gamma_4\rangle\) and \(|4\rangle = |\Gamma_2 - i\Gamma_4\rangle\), which are exactly along either \([110]\) or \([110]\) direction (ensured by the \(C_{2v}\) symmetry) and also real simultaneously (ensured by the time-reversal symmetry). An effective \(2 \times 2\) Hamiltonian can be constructed from these two bright states \([12]\),

\[
H = \tilde{E} + \delta \sigma_z + \kappa \sigma_x,
\]

(1)

where \(\tilde{E} + \delta = \langle 3 | H | 3 \rangle\), \(\tilde{E} - \delta = \langle 4 | H | 4 \rangle\), \(\kappa = \langle 3 | V_1 | 3 \rangle\), and \(\delta, \kappa \in \mathbb{R}\). \(\sigma_z\) and \(\sigma_x\) are Pauli matrices. \(\tilde{E}\) defines the exciton energy, and \(\Delta = 2\sqrt{\kappa^2 + \delta^2}\) defines the FSS. Physi-
nally, $\kappa$ describes the coupling between two bright states, leading to the deviation of the emission line from $[110]$ and $[\overline{1}0\overline{1}]$ directions. The wavefunction of the bright exciton can be written as $\psi = \cos(\theta)|3\rangle + \sin(\theta)|4\rangle$, where $\theta$ is the polarization angle with $\tan(\theta) = \kappa^{-1}(\kappa \pm \sqrt{\delta^2 + \kappa^2})$. Hence $\delta = \pm \Delta \sin(2\theta)/2$, and $\kappa = \mp \Delta \cos(2\theta)/2$.

The $V_1$ term can be uniquely determined with the following recipe. We assume $V$ is the total potential of QDs (including all types of interactions), and $G$ contains all irreducible representations of $C_{2v}$ point group. For any $g \in G$, $gH_{2v}g^{-1} = H_{2v}$, therefore $H_{2v} = T + \langle \sum_{g \in G} gV(r)g^{-1} \rangle/|G|$, where $T$ is the kinetic energy and $|G|$ is the number of symmetry operators. Then

$$V_1(r) = V(r) - \langle \sum_{g \in G} gV(r)g^{-1} \rangle/|G|. \tag{2}$$

Obviously, $\sum_{g} gV_1(r)g^{-1} \equiv 0$ for any $r$. The $C_{2v}$ symmetry ensures the direct connection between $r$ and $grg^{-1}$, whereas there are no correlations for other coordinate pair $(r, r')$ when $r' \neq grg^{-1}$. Thus the $V_1$ term, which depends essentially on the local details of QDs, has the basic feature even in a single QD that the potential should be spatially changed rapidly both in sign and magnitude.

The major difference between single QD and QDE is that in the latter case the morphology variations of QDs make $H_{2v}$, and $V_1$ also varying. To make the physical picture more transparent, we rewrite $H_{2v} = \bar{H}_{2v} + \delta H_{2v}$, where $\bar{H}_{2v} = \langle H_{2v}\rangle$, and $\delta H_{2v}$ defines the variations from QD to QD. Now, $V_1$ can be treated as an random potential from the viewpoint of QDEs due to its peculiar feature in Eq. 2. The $\delta H_{2v}$, although lacking similar feature, also exhibits some degrees of randomness. We propose that the effective model in Eq. 1 for a single QD is still valid to describe the optical properties of QDEs, with the modification that $\delta$ and $\kappa$ be treated as independent random numbers satisfying some particular distributions. Notice that both $\delta H_{2v}$ and $V_1$ contribute to the randomness of $\delta$, while only $V_1$ contributes to $\kappa$. We expect $\langle \kappa \rangle = 0$, but $\langle \delta \rangle \neq 0$ because QDs with $C_{2v}$ symmetry ($V_1 \equiv 0$) still have finite FSS. We set $\delta = \delta_0 + \delta'$, where $\delta_0 = \langle \delta \rangle$. In the following, it is essential to verify the randomness of $\delta$ and $\kappa$ to validate our basic effective model. We will also show that $\delta_0$ characterizes the shape anisotropy effect of the QDEs.

Intuitively, the distributions of FSS and polarization angle should depend strongly on the morphology details of QDs, including size (base diameter, height), shape, alloy profile, etc. However, it is impossible to quantitatively determine all these parameters in experiments. We circumvent this difficult by defining several physical parameters to fully characterize the statistical features of FSS and polarization angle in QDEs.

The distribution of any observable physical quantity, say $f = f(\kappa, \delta_0, \delta')$, is defined as

$$P(z) = \int d\delta' d\kappa d(f - z)N(\delta', \sigma_\delta)\mathcal{N}(\kappa, \sigma_\kappa). \tag{3}$$

where $\kappa$ and $\delta'$ satisfy normal distributions $\mathcal{N}(x, \sigma)$, with variations $\sigma_\kappa = \langle \kappa^2 \rangle$ and $\sigma_\delta = \langle (\delta')^2 \rangle$, respectively. The exact distribution functions for FSS and polarization angle can be found in the supplementary material.

The advantage of our strategy is that we are able to define and characterize the statistical features of FSS and polarization angle of the QDEs without knowing the morphology details of the samples.

Here, we are particularly interested in the distribution of FSS with $\delta_0 = 0$, which corresponds to the results of lens-shaped QDEs. The distribution of FSS reads as

$$P(\Delta) = \frac{\Delta}{4\sigma_\delta \sigma_\kappa} \exp(-A_+ \Delta^2)I_0(A_- \Delta^2), \tag{4}$$

where $A_\pm = 1/(16\sigma_\delta^2) \pm 1/(16\sigma_\kappa^2)$ and $I_0(x)$ is the modified Bessel function of the first kind. We find that for a large class of QDEs the distribution of FSS can be well approximated by the Wigner function

$$P(s) = \frac{\pi}{2} \exp(-\frac{\pi}{4} s^2), \tag{5}$$

FIG. 1: (Color online). (a) - (d) distribution of $\delta$ (■) and $\kappa$ (■) for different QDEs. The solid lines and dashed lines are the best fitting to Gaussian distributions, and the fitted parameters are tabulated in Table I. The exciton energy dependence of $\delta$ and $\kappa$ are plotted in (e) and (f), respectively.
worth emphasizing that the fluctuation of \( V \) the random coupling between the bright states from parameter equations will also be examined. We study several rather different QDEs here to demonstrate the strong repulsion between levels. More specifically, the strong repulsion is related to the independence of off-diagonal and diagonal elements, which leads to the appearance of the first \( s \) term in (5). It is exactly this term that renders the probability of finding QDs with vanishing FSS very small. It therefore should be in stark contrast to the trivial random matrix without off-diagonal elements where strong attractive between levels leads to level spacings described by Poisson distribution (2).

The distribution of the polarization angle \( \theta \) in \([-\pi/2, \pi/2]\) at \( \delta_0 = 0 \) reads as

\[
P(\theta) = \frac{1}{\pi} \frac{1}{\eta \sin(2\theta)^2 + \eta^{-1} \cos(2\theta)^2},
\]

where \( \eta = \sigma_\delta / \sigma_\kappa \). We see \( P(\theta) = 1/\pi \) when \( \eta = 1 \). \( P(\theta) \) reaches the maximum at \( \theta = 0, \pm \pi/2 \) when \( \eta > 1 \), or at \( \theta = \pm \pi/4 \) when \( \eta < 1 \). The period of \( P(\theta) \) is \( \pi/2 \). It is worth emphasizing that the fluctuation of \( \theta \) is induced by the random coupling between the bright states from \( V_1 \), thus it is not an extrinsic effect [33]. In the following, we will confirm the validity of the above analytical results using several rather different QDEs, some of which even have relative large \( \delta_0 \). The validity of the above one-parameter equations will also be examined.

In the limit of \( \delta_0 \gg \sigma_\delta, \sigma_\kappa \), the statistical features turn out to be very simple and can be understood as follows. The polarization angle should be around either \( \theta = 0 \) or \( \theta = \pi/2 \), thus \( \eta \gg 1 \). The FSS is most likely to be observed at \( \Delta \sim \delta_0 \) in such types of QDEs, and the distribution of FSS decays rapidly to zero when \( |\Delta - \delta_0| \gg \sigma_\delta \). The distribution of FSS is more close to a Gaussian function with width \( \sqrt{\sigma_\delta^2 + \sigma_\kappa^2} \). This limit actually corresponds to QDEs with strong shape anisotropy.

**Theoretical and experimental verifications.** Above analytic results are further confirmed using atomistic simulation and realistic experiments. The simulation is based on the well-established atomistic pseudopotential method [32,33]. We consider three different In\(_x\)Ga\(_{1-x}\)As/GaAs QDEs: (T1) Lens QDs with fixed \( x = 0.6 \), diameter \( D = 25 \) nm, and height \( h = 3.5 \) nm; (T2) Elongated QDs with \( x = 0.6 \), diameter along [110] (25 nm), \( D_{[110]} = 26 \) nm and \( D_{[\bar{1}10]} = 24 \) nm, \( h = 3.5 \) nm; (T3) Lens QDs where \( x \), \( D \) and \( h \) are variables, with the mean values \( \langle x \rangle = 0.6 \), \( \langle D \rangle = 25 \) nm, \( \langle h \rangle = 3.5 \) nm, and variations up to 10\% of the corresponding mean values. In the experiments we consider four different QDEs. The samples were grown by solid source molecular beam epitaxy on GaAs (001) substrates. GaAs/AlGaAs QDs are obtained by infilling self-assembled nanoholes fabricated in situ either by droplet etching [35] or by selective AsBr\(_3\) etching [36]. The indium flushed self-assembled In(Ga)As/GaAs QDs are grown by Stranski-Krastanov technique [37]. Excitons confined in the GaAs QD (QDE E1), In(Ga)As QD (QDE E4) and GaAs quantum well potential fluctuations (QDEs E2 and E3, respectively) are investigated. All the microphotoluminescence spectroscopy measurements were performed at low temperature. We study several rather different QDEs here to show the validity of the generic model. In the following, we choose QDEs T1, T2, E1 and E2 to present our major findings, while all the parameters for the QDEs, including the fitted results, are summarized in Table I.
TABLE I: Summarized parameters for different QDEs. The definition of QDs can be found in the main text and supplementary material [28]. N the sample volume of simulated or measured QDs with $E_X$ (eV) the mean exciton energy, and $\sigma_X$ (meV) the variation of the exciton energies. ($\kappa$), $\sigma_\kappa$, $\delta_0$, $\delta_\kappa$, $\langle \Delta \rangle$ (the mean value of FSS) are all in unit of meV. $\eta$ has been defined in Eq. 6 $G^{(2)} = \langle (\delta - \bar{\delta})(\kappa - \bar{\kappa}) \rangle / \sigma_\kappa \sigma_\kappa$ measures the cross correlation between the two random numbers. $P$ (%) defines the probability of finding QDs with FSS smaller than the broadening of the emission line (1 muV).

| QDs      | T1   | T2   | T3   | E1   | E2   | E3   | E4   |
|----------|------|------|------|------|------|------|------|
| $N$      | 1351 | 1381 | 7714 | 204  | 412  | 401  | 240  |
| $E_X$    | 1.240| 1.210| 1.186| 1.158| 1.722| 1.768| 1.386|
| $\sigma_X$| 2.9  | 3.0  | 28.3 | 2.5  | 3.6  | 5.1  | 4.4  |
| $\langle \kappa \rangle$| -0.1 | -0.2 | -0.2 | -0.1 | 0.2  | -0.3 | -0.4 |
| $\delta_0$| 1.7  | 2.0  | 1.5  | 2.1  | 2.3  | 1.3  | 4.4  |
| $\delta_\kappa$| 1.4  | 1.8  | 1.3  | 3.0  | 13.6| 10.2| 4.8  |
| $\langle \Delta \rangle$ | 4.0 | 7.7 | 3.8 | 32.6 | 39.7 | 15.4 | 11.7 |
| $\eta$   | 0.88 | 1.99 | 1.04 | 7.80 | 7.53 | 5.26 | 1.0  |
| $G^{(2)}$| 0.002| 0.003| ~0.0 | -0.041| -0.298| 0.114| -0.08 |
| $P$      | 5.0  | 1.3  | 5.7  | ~0.0 | 0.2  | 0.1  | 0.5  |

For all QDEs we observe that the distributions of $\delta$ and $\kappa$ (see Fig. 1a-d) can be well fitted with Gaussian functions. We have also confirmed that these random variables are independent of exciton energies (see Fig. 1f, g). In QDE T1, we find $\delta_0 \sim 0$ and $\sigma_\kappa \simeq \kappa$, see Fig. 1h, which agree well with the experimental data in QDE E4; In elongated QDE T2 (Fig. 1b), the shape anisotropy leads to significant nonzero $\delta_0$. This observation qualitatively agrees with the results in experiments, see Fig. 1d, which have elongation either along [110] or [110] direction. To verify the randomness of these two parameters, we calculate the cross correlations between $\delta'$ and $\kappa$ and find that the correlation between them is indeed very small, see $G^{(2)}$ in Table I. The experimental measured correlation is larger than our atomistic simulation because much smaller sample volume is measured in experiments. The fluctuations of $\delta$ and $\kappa$ both in simulations and experiments are found to be of the order of several muV; $\langle \kappa \rangle \sim 0$ is also consistent with expectation.

The connection between morphology and FSS can be established as follows. We observe that the anisotropic effect arising from the lattice nonequivalence of [110] and [110] directions in zinc-blende crystals leads to a fairly small $\delta_0$, whereas the anisotropic effect arising from shape elongation generally leads to a significant $\delta_0$ (and hence large FSS), as observed in QDE T2 and the experimental samples. In experiments, the QDs exhibit apparent elongation either along [110] or [110] direction, and for the quantum well potential fluctuation QDE, strong shape anisotropic effect is also expected. The basic conclusion that shape anisotropy has dominate contribution to FSS is consistent with the recent reports by Plumhof [20] and Huo [28]. However what we advance here is that the shape anisotropy can be fully characterized by $\delta_0$. We note that the morphology of a single QD is impossible to be precisely controlled in experiments. However, the statistical properties of QDEs maybe well controlled, therefore it provides an interesting arena in future to study the relationship between these parameters and the growth environments, such as temperature, pressure, etc.

The distributions of FSS for QDEs are presented in Fig. 2. The dashed lines are the best fitting using Eq. 5 while the dot-dashed lines are calculated from Eq. 6 with the three parameters obtained directly from fitting the distributions of $\delta$ and $\kappa$ with Gaussian distributions (see Fig. 1). We have numerically confirmed that the distribution of FSS can be well described using Eq. 5 when $\delta_0 < 2\sigma_\delta$, and it is somewhat poorer for QDEs with large $\delta_\kappa/\sigma_\kappa$ (see QDE E1). The distributions of polarization are presented in Fig. 3 and generally much better agreement can be obtained. The good agreement between analytical curves from Eq. 5 and simulations/experiments clearly demonstrate the validity of our model. Here we deliberately verify the validity of Eq. 5 and 6 to the condition $\delta_0 \neq 0$ to provide important reference for future researches in QDs and other nanostructures.

Discussions and concluding remarks.- We now answer the three fundamental questions put forward in the introduction. Firstly, the probability of finding QDs with FSS smaller than 1 meV is $P = 1 / 16\delta_\sigma \sigma_\kappa$ when $\delta_0 = 0$. For typical values of $\sigma_\sigma$ and $\sigma_\kappa$, $P \sim 1\%$. When $\delta_0 \neq 0$, a prefactor $\exp(-\delta_0^2/2\sigma_\delta^2)$ arises, which further suppresses $P$, see Table I (Q:I). Secondly, in a random potential $V_1$, it is possible to observe two QDs with the same exciton energies and the same FSSs, but fairly different polarization angles. No obvious correlation between FSS and polarization angle can be derived (Q:II). Finally, due to the randomness of $\delta$ and $\kappa$, the morphology effect can be missed out in experiments of single QD [10, 11]. However, it can be recovered from experiments about QDEs. In particular, for QDE with a large FSS ($\delta_0 \gg \sigma_\kappa$, $\kappa^\prime$), the mean value of FSS itself can be used to characterize the shape anisotropy effect of QDE, in which condition the emissions should almost polarized along either [110] or [110] direction (Q:III).

Several additional remarks are in order. Firstly, the basic idea can be easily generalized to study the optical properties of high-symmetric QDEs. Although the two bright states are degenerate for QDs with $C_{3v}$ or $D_{2d}$ symmetry [17], the random term $V_1$ can still render the probability of finding QDs with vanishing FSS small, as seen in recent experiments [11, 12]. For high-symmetric QDEs, $\delta_0 = 0$, thus only two independent parameters are required to fully characterize the statistical properties of FSS and polarization angle. We estimate $\sigma_\kappa$ and $\sigma_\delta \sim 1$ meV using the results from Ref. 11, which seems to a bit smaller than that in $C_{3v}$ symmetric QDEs, see.
Table I. Secondly, the effective model is derived purely from symmetry argument, and is independent of the morphology details of QDs, thus it is also applicable to study the optical properties of other semiconductor nanostructures, e.g., quantum rod and colloid nanocrystals. As a generic feature, all the physical observations should exhibit some degree of random fluctuations. Thirdly, the leaning from QDE requires the photons have good polarization property, and recently there are indeed some remarkable progresses along this line. The statistical feature of polarization angle can find important application in these fields. Finally, since the random potential $V_1$ is impossible to be captured by any theoretical model, the theoretical modeling can only be used to qualitatively, instead of quantitatively, interpret the physical observations in experiments. To conclude, all these results and insights yield a completely new physical picture to understand the optical properties of QDEs.

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