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## **Room Temperature Anomalous Coherent Excitonic Optical Stark**

## **Effect in Metal Halide Perovskite Quantum Dots**

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**ABSTRACT.** Non-resonant optical driving of confined excitons and biexcitons in low dimensional semiconductors can open up exciting opportunities for experimentally realizing strongly interacting photon-dressed (Floquet) states through optical Stark effect (OSE) for coherent modulation of exciton state. Here, we report the first room temperature observation of Floquet biexciton mediated anomalous coherent excitonic OSE in CsPbBr3 quantum dots (QDs) using circularly polarized ultrafast lasers. Remarkably, the strong exciton-biexciton interaction leads to the coherent redshift and splitting of the exciton resonance as a function of the drive photon frequency similar to Autler-Townes splitting in atomic and molecular systems. The large biexciton binding energy of  $71 \pm 1$  meV and exciton-biexciton transition dipole moment  $25 \pm 1$  Debye makes it possible to observe the hallmark results even at large detuning energies of more than 300 meV. This is accompanied by an unusual crossover from linear to non-linear fluence dependence of the OSE as a function of drive photon frequency. Our systematic studies in the metal halide perovskite QDs reveal crucial information on the unexplored many-body coherent interacting regime, making them suitable for room temperature quantum devices based on spin qubits.

Time-periodic optical driving is a powerful tool for manipulating quantum systems and has led to exotic quantum phases such as time crystals<sup>1</sup> and topological phases<sup>2,3</sup> that will benefit quantum metrology<sup>4</sup>. Of particular interest is the excitonic optical Stark effect (OSE) commonly described as the energy shift in a two-level system arising from the coherent nonlinear light-matter interactions<sup>5–9</sup>. This can be understood in terms of state repulsion of the photon-dressed states (Floquet states) and the original states that exist only for the coherent (pump pulse) duration. In general, the spectroscopic evidence of the OSE exhibits a blueshift of the transition energy<sup>5,7,10–14</sup>, which increases linearly with the light intensity and inversely with the detuning energy. Multiple studies in bulk semiconductors<sup>11</sup> and nanostructures (0-2 dimension)<sup>5,10,12,13,15,16</sup> have explored the nature of spin-selective excitonic OSE for a non-resonant photoexcitation in the framework of a non-interacting exciton regime. These studies demonstrated a strong coherent signal resulting from blueshift of exciton absorption only for the same pump-probe polarization. Because of the coherent nature, OSE has many promising applications in engineering the quantum states <sup>13,17,18</sup> and coherent optical modulation for switching<sup>18–20</sup> and generating mode-locked pulses<sup>18,21,22</sup>. In contrast, the interacting regime of exciton-biexciton coupling leads to the phenomena that are qualitatively different from those in the non-interacting exciton framework and result in the coherent redshift of the exciton resonance. Optical Stark redshift has been observed in CuCl single crystal<sup>23,24</sup>, monolayer transition metal dichalcogenides<sup>17,25</sup>, InGaAs quantum wells<sup>26</sup>, and molecular beam epitaxy grown single QDs like Mn doped CdTe<sup>27</sup>, InGaAs/GaAs<sup>28</sup>, and GaAs/AlGaAs<sup>29</sup> having appreciable exciton-biexciton coupling. Such effects are limited to only cryogenic temperatures mainly because of the low binding energies of the biexcitons which is of the order of several tens of µeV to less than several tens of meV (< 40 meV). Obviously, for many practical applications

involving quantum states of matter, it would be desirable to observe these effects at room temperature.

Metal halide perovskite (MHP) quantum dots (QDs) of the general formula ABX<sub>3</sub> (where A = MA/FA/Cs; B = Pb/Sn; and X = Cl/Br/I) are particularly attractive for studying the coherent light-matter interactions since in many ways they behave as a molecular system of customizable atoms like features<sup>30–33</sup>. The low-cost solution processability and easiness to integrate with existing nanofabrication facility make them much more attractive<sup>34,35</sup>. Moreover, the strong band-edge exciton-photon coupling<sup>36</sup>, defect tolerant electronic structure of trap-free nature<sup>34,35,37–39</sup>, discrete and narrow-bandwidth exciton transitions<sup>5,11,39</sup>, and scalable production of highly coherent quantum emitters<sup>37,40</sup> make them an ideal candidate to study the coherent and spin selective lightmatter interactions at room temperature. Not to mention, the multiexciton interactions in MHP QDs, particularly CsPbBr<sub>3</sub>, is reasonably strong<sup>32,41</sup> and thus can influence the coherent lightmatter interactions. Recently, we have demonstrated a spectrally well-resolved exciton and biexciton resonance in a weakly confined CsPbBr<sub>3</sub> QDs with a high binding energy ~30-40 meV for both excitons and biexcitons at room temperature 42,43. The biexciton binding energy can further easily be tuned to higher values using quantum confinement effect (reducing the QD size)<sup>32</sup>. Such control over the biexciton binding energy is much more difficult in epitaxially grown QDs. In line with these expectations, OSE in MHP QDs are often overlooked by existing experimental and theoretical models<sup>5,11,12</sup>, which question the nature of coherent exciton-exciton interactions in a two level model. Despite possessing favourable properties and being a potential candidate, a complete understanding of the coherent OSE in strongly interacting excitons framework in MHP QDs at room temperature is lacking and largely remains unexplored. Here, using solution processed strongly confined CsPbBr<sub>3</sub> QDs  $\sim$  6 nm having biexciton binding energy 71  $\pm$  1 meV,

we demonstrate the first room temperature experimental realization of the biexciton mediated anomalous OSE, which evolves from the energy redshift to the splitting of the exciton resonance as a function of the drive photon frequency. This is accompanied by the unusual crossover from linear to nonlinear fluence dependence of the optical Stark shift. The unusualities observed stimulated the consideration of the three-level system than just the two level system commonly employed in non-interacting framework. By constructing an extended Jaynes-Cummings model for three-level system we could account for all the main observations in our experiment. These findings reveal the strong influence of exciton-biexciton coupling on the coherent modulation of the exciton resonance in MHP QDs. Further the Rabi energy determined to be  $83 \pm 2$  meV and corresponding exciton-biexciton transition dipole moment  $25 \pm 1$  Debye are larger than any known systems, promoting the strong candidature of MHP QDs for exploitation in applications involving many-body coherent phenomena such as entangled photon pair generation and biexciton lasing.

Long-chain, zwitterion-capped CsPbBr<sub>3</sub> QDs were synthesized by facile hot injection method, and spin-coated onto an ITO-coated glass microscope slide for optical measurements; the details are provided in the Supporting Information. A high angle annular darkfield scanning transmission electron micrograph (HAADF-STEM) (Figure 1a) confirm the high degree of size uniformity and a cuboid morphology with an average size of 6 nm that lies in the region of strong quantum confinement. The ground-state optical absorption of the sample shown in Figure 1b demonstrates a sharp exciton peak and a continuum band. By using the Elliott model for the Wannier-Mott exciton  $^{44,45}$ , details in Supporting Information, we calculated the exciton resonance energy (E<sub>0</sub>) =  $2.405 \pm 0.002$  eV and continuum energy  $2.479 \pm 0.002$  eV, which reveal the exciton binding energy,  $E_x = 74 \pm 2$  meV, that agrees with our temperature-dependent photoluminescence (PL) measurements discussed in the Supporting Information.

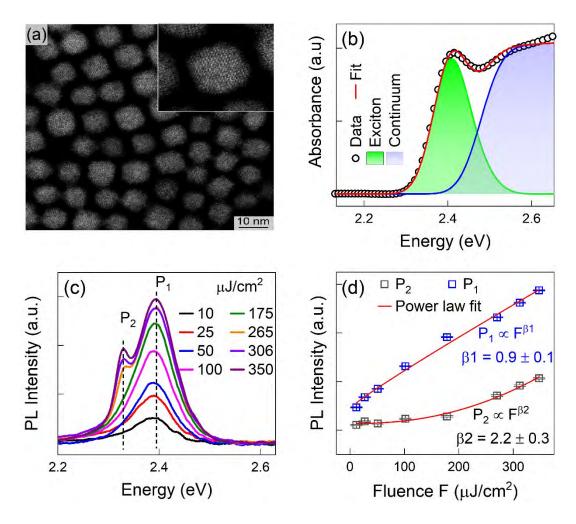


Figure 1. Morphological and optical characterization of CsPbBr<sub>3</sub> QDs. (a) HAADF-STEM of a typical sample of ca. 6 nm CsPbBr<sub>3</sub> QDs; (b) Optical absorption spectrum fitted using the Elliott model; (c) Intensity dependent PL demonstrating exciton ( $P_1$ ) and biexciton ( $P_2$ ) emission; (d) Power-law fitting of  $P_1$  (~2.39eV) and  $P_2$  (~2.32 eV) intensity revealing a near-linear and near-quadratic intensity dependence with exponent  $\beta 1$  ~0.9 and  $\beta 2$  ~2.2 indicating that it is due to exciton and biexciton emission, respectively.

To identify the biexciton states, we studied the PL under 400 nm pulsed excitation with different excitation fluences (Figure 1c), details in Supporting Information. At low excitation fluence, we observed PL from exciton emission (P<sub>1</sub>), however, a second sharp peak (P<sub>2</sub>) emerges towards the low energy at higher fluences. The P<sub>1</sub> peak exhibit a near linear fluence dependence

 $(I_{P1} \propto Fluence^{\beta 1} \text{ with } \beta 1 = 0.9 \pm 0.1)$  as expected from the exciton emission peak. On the other hand, the superlinear fluence dependence  $(I_{P2} \propto Fluence^{\beta 2} \text{ with } \beta 2 = 2.2 \pm 0.3)$  of  $P_2$  shown in Figure 1d reveals that it is originating from the biexciton transition<sup>46–48</sup>. The details are given in Supporting Information. Strikingly, above a threshold pump fluence of  $175 \,\mu\text{J/cm}^2$ , we observed amplified spontaneous emission kind of behaviour at the biexciton position with a reduced full-width half maximum. The biexciton binding energy  $E_b$  calculated using  $P_1$ - $P_2$  =  $71 \pm 1 \,\text{meV}$ , is well in agreement with the  $E_b$  obtained from our transient absorption measurements, details in the Supporting Information. The Haynes factor,  $E_b/E_x$  =  $0.96 \pm 0.05$ , is consistent with our recent report in weakly confined CsPbBr<sub>3</sub> QDs<sup>42</sup> and is an order of magnitude larger than the bulk and five times the 2-dimensional and quantum well semiconductors, demonstrating a very robust correlation of excitons<sup>48–50</sup>.

We employed circularly polarized transient absorption spectroscopy to measure the interacting many-body Floquet state mediated OSE in strongly confined CsPbBr<sub>3</sub> QDs. An optical parametric amplifier generating ~120 fs broadly tunable 2.08-2.32 eV pump pulses was used to excite the sample. The resulting changes in the absorption spectrum ( $\Delta A$ ) were measured using a probe pulse in the photon energy range 2.30–2.55 eV. The experimental setup details are provided in the Supporting Information. To generate the coherent OSE and avoid any real population excitation, the pump photon energy was kept below E<sub>0</sub>. Figure 2a,b show a compilation of the room temperature transient absorption spectra for  $\sigma$ +pump and  $\sigma$ -probe ( $\sigma$ + $\sigma$ -) and  $\sigma$ + $\sigma$ + for detuning energies ( $\Omega$  =E<sub>0</sub>- $\hbar\omega$ ) 316 and 82 meV at a fixed pump pulse fluence of 1 mJ/cm<sup>2</sup>, where  $\hbar\omega$  is pump photon energy. Here,  $\sigma$ + and  $\sigma$ - denote right circular and left circular polarization, respectively. The top panel presents the pseudo-color plot of the  $\Delta A$ , and the bottom panel represents the cross-section close to zero pump–probe delay. The instantaneous response of the

 $\Delta A$  spectrum shows that the signal is primarily originating from the coherent OSE. First, we discuss the  $\Omega = 316$  meV  $\Delta A$  spectrum for  $\sigma^+ \sigma^-$  close to zero pump-probe delay.

The spectrum manifests a bleach at  $E_0$  and the emergence of an induced absorption (2.40 eV) appearing below  $E_0$ , displaying a 5 meV coherent redshift of the exciton transition. We used the traditional Gaussian shifting method<sup>43,44</sup> to extract the Stark shift from the experimental data. Our results are compelling and show a sharp contrast with the previous studies on bulk and two-dimensional MHP films, where no signal for bulk<sup>11</sup> and weaker transient signal for two-dimensional MHPs<sup>12</sup> was observed for the  $\sigma^+\sigma^-$ .

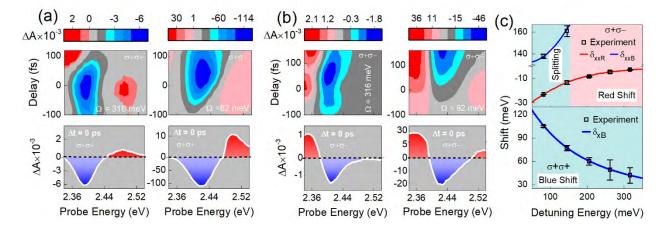


Figure 2. Anomalous excitonic OSE at fluence of  $1mJ/cm^2$ . Pseudo-colour plot (top) and crossection at near zero pump-probe delay (bottom) at detunings of 316 (left) and 82 meV (right) with pump fluence of  $1mJ/cm^2$  for (a)  $\sigma^+\sigma^-$  and (b)  $\sigma^+\sigma^+$ . The existence of a strong signal only for the pump-pulse duration confirms the coherent response. However, at longer delays, i.e., after  $\sim 120$  fs, a weak signal also exists due to the unintentional real-excitation population arising from two-photon absorption; (c) The stark shift evaluated from the experimental data (symbols) showing an excellent agreement with theoretical calculations (solid lines).

This coherent redshift clearly demonstrates that the non-interacting exciton picture cannot fully describe the OSE in CsPbBr<sub>3</sub> QDs since they have a strong biexciton state with  $E_b \approx E_x$  at room

temperature. To get further information, we have analyzed the  $\Delta A$  spectrum by continuously reducing  $\Omega$  from 316 to 82 meV, Figure S5 in Supporting Information. Strikingly, for  $\Omega = 82$  meV, the spectrum is dominated by the exciton energy splitting of Autler-Townes type with induced absorption at energies both above and below  $E_0$  and a reduction of bleach at  $E_0$  as shown in Figure 2a. This anomalous OSE of exciton shift originates from the remarkably high biexciton binding energy that embarks the observation of hallmark results even at large detuning energies of more than 300 meV, which is three orders of magnitude larger than CuCl single crystals<sup>24</sup>, around two orders of magnitude larger than InGaAs quantum wells<sup>26</sup> and InGaAs/GaAs single QD<sup>28</sup>, and four times of monolayer transition metal dichalcogenides<sup>17</sup>. Our experimental results demonstrate the importance of many-body interactions determining the OSE in MHP QDs.

The  $\sigma^*\sigma^+$   $\Delta A$  spectrum of CsPbBr<sub>3</sub> QDs for  $\Omega$  = 316 meV close to zero pump–probe delay indicates a 40 meV coherent blueshift of the exciton absorption, manifested as a reduction in  $E_0$  and the emergence of an induced absorption at 2.445 eV (Figure 2b). The blueshift is present there for all  $\Omega$  and becomes more significant as pump energy approaches exciton resonance, Figure 2b and Figure S5 in the Supporting Information, similar to the ordinary OSE previously reported in MHPs in a non-interacting exciton regime<sup>5,11,12</sup>. It is important to note here that despite the below bandgap excitation, we cannot discard the possibilty of the presence of real population of the carriers, since the excitation fluence is sufficiently high to cause the two photon absorption (TPA) that result in the hot carrier population in the system. The remanent signal after the coherent duration shown in Figure S6 in the Supporting Information, manifest the signal originating from TPA induced real carrier population. However, this will not affect the main result of biexciton mediated coherent OSE presented in this manuscript. The TPA will create the hot carrier population in the system, and these hot carriers will take time to travel to the band-edge and by

that time the signal originating from the coherent interactions will be over. To experimentally support our claim, we plot the amplitude of the coherent bleach in  $\sigma^+\sigma^-$  at zero pump-probe delay against the fluence in the Supporting Information, Figure S7. The figure manifests the linear fluence dependence in stark contrast to the quadratic dependence expected from the TPA induced signal<sup>12</sup>, confirming that the coherent signal is independent of TPA. To further support our argument, first, we measure the threshold fluence (> 0.5 mJ/cm<sup>2</sup>) for TPA at  $\hbar\omega = 2.32$  eV using a conventional Z-scan and then measure the OSE below the threshold fluence (~ 0.3 mJ/cm<sup>2</sup>), Figure S7 in Supporting Information. The Autler-Townes like splitting of the exciton resonance even at this low fluence implies that the OSE observed here is originating from the coherent interactions and not from the TPA.

To uncover further experimental facts promoting the idea of interacting exciton many-body states in the OSE of CsPbBr<sub>3</sub> QDs, we measured the optical Stark shift as a function of  $\Omega$  (Figure 2c). The exciton energy redshift well defined away from the exciton–biexciton resonance for  $\sigma^+\sigma^-$  evolve to an energy splitting of Autler-Townes type as  $\Omega$  approaches to resonance. For  $\sigma^+\sigma^+$ , we did not observe any such split of the exciton resonance. Strikingly, the Autler-Townes like exciton splitting for  $\sigma^+\sigma^-$  increases with increase in  $\Omega$ , in stark contrast to decrease in blueshift for  $\sigma^+\sigma^+$ , describing the unusual OSE in terms of  $\Omega$ .

To understand the mechanism behind the anomalous OSE, we took into consideration of the exciton-biexciton coherent interaction and constructed an extended Jaynes-Cummings model for three-level<sup>51,52</sup>, as shown in Figure 3a-c, details in the Supporting Information. In the figure,  $|g, n\hbar\omega\rangle$ ,  $|x, n\hbar\omega\rangle$ ,  $|x', n\hbar\omega\rangle$ , and  $|xx', n\hbar\omega\rangle$  represent bare light-matter states that account for exciton and biexciton absorption. Here  $|g\rangle$ ,  $|x\rangle$ ,  $|x'\rangle$ , and  $|xx'\rangle$  represent the ground state, spin-up exciton, spin-down exciton, and biexciton, respectively. The integer n represents the number of

photons at the pump frequency  $\omega$ . This three-level model, used previously to quantify coherent modulation of the quantum states in semiconductors<sup>53–56</sup>, can easily be modified in the presence of exciton-biexciton interactions to describe the anomalous OSE in CsPbBr<sub>3</sub> QDs.

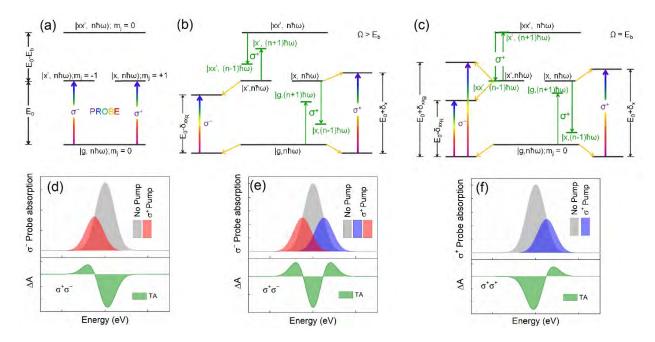


Figure 3. Three-level Jaynes-Cummings model. (a) illustration of probe absorption in absence of pump; Floquet states (green lines) and resulting shifts under  $\sigma^+$  pump for (b)  $\Omega > E_b$  and (c)  $\Omega \approx E_b$ . The red and blue arrows represent the redshift and blueshift of the exciton resonance, respectively; (d)-(f) Illustration of coherent signal arising on (d) redshift and (e) splitting under  $\sigma^+\sigma^-$ , and on (f) blueshift under  $\sigma^+\sigma^+$ .

In our scheme, the  $\sigma^+$  probe measures the change in  $|g, n\hbar\omega\rangle \rightarrow |x, n\hbar\omega\rangle$  transition, while  $\sigma^-$  measures the change in  $|g, n\hbar\omega\rangle \rightarrow |x', n\hbar\omega\rangle$  in presence of an intense non-resonant  $\sigma^+$  pump, detuned below the exciton resonance energy, producing a series of photon replicas of the original states known as the Floquet states (green lines), Figure 3a-c. Floquet states having the same  $m_j$  (magnetic quantum number) as that of original states repel each other (Figure 3b,c) giving rise to

the novel coherent features. With this idea, we first constructed the effective Hamiltonian (H) for the  $\sigma^+\sigma^-$  taking  $|g, n\hbar\omega\rangle$ ,  $|x, (n-1)\hbar\omega\rangle$ ,  $|x', n\hbar\omega\rangle$ , and  $|xx', (n-1)\hbar\omega\rangle$  as basis states described by

$$H = \begin{pmatrix} 0 & V_x & 0 & 0 \\ V_x & \Omega & 0 & 0 \\ 0 & 0 & E_0 & V_{xx} \\ 0 & 0 & V_{xx} & E_0 + \Omega - E_b \end{pmatrix}$$
 (1)

Where  $V_x = \mu_x E_{effective}$  and  $V_{xx} = \mu_{xx} E_{effective}$  are the Rabi energies which are proportional to square root of fluence.  $E_{effective}$ ,  $\mu_x$ , and  $\mu_{xx}$  describe the pump photon electric field, exciton transition dipole moment, and exciton-to-biexciton transition dipole moment, respectively. In general,  $\mu_{xx} \approx \mu_x^{57}$  and so  $V_{xx} \approx V_x$ . The Eigen values of the Hamiltonian matrix represent the redshift  $(\delta_{xx_R})$  and blueshift  $(\delta_{xx_R})$  of the exciton resonance as

$$\delta_{\chi\chi_R} = \frac{\sqrt{(\Omega - E_b)^2 + 4V_{\chi\chi}^2}}{2} - \frac{\Omega - E_b}{2} - \frac{\sqrt{\Omega^2 + 4V_\chi^2}}{2} + \frac{\Omega}{2}$$
 (2)

$$\delta_{xx_B} = \frac{\sqrt{(\Omega - E_b)^2 + 4V_{xx}^2}}{2} + \frac{\Omega - E_b}{2} + \frac{\sqrt{\Omega^2 + 4V_x^2}}{2} - \frac{\Omega}{2}$$
(3)

These equations show that the exciton–biexciton interactions dramatically modify the coherent light–matter interactions from the non-interacting exciton picture discussed in previous studies  $^{5,11,12}$ , and lead to a new set of optically driven coherent phenomena as a function of  $\Omega$ . The detailed description is given in the Supporting Information. In particular, depending upon whether  $\Omega > E_b$  or  $\Omega \approx E_b$ , the |xx'|,  $(n-1)\hbar\omega$  lies above or nearly at the same level of |x'|,  $n\hbar\omega$  (Figure 3b,c), respectively resulting in redshift or splitting of the exciton state. Noticeably, in both cases the |x|,  $(n-1)\hbar\omega$  lies above the |g|,  $n\hbar\omega$  resulting in the downshift of the ground state. The exact shift in exciton resonance is the combined shift in ground and exciton state (Equation 2 and 3), details in the Supporting Information. The schematic of coherent transient signal resulting from redshift and splitting of exciton resonance is shown in Figure 3d,e.

After demonstrating the anomalous OSE, we set up the effective Hamiltonian matrix for the  $\sigma^+\sigma^+$  in the interacting exciton picture as discussed in the Supporting Information and the eigen values of the matrix correspond to the blueshifted excitonic level is given by

$$\delta_{\mathbf{x}} = -\Omega + \sqrt{\Omega^2 + 4V_{\mathbf{x}}^2} \tag{4}$$

Since the  $|g, (n+1)\hbar\omega\rangle$  lies below the  $|x, n\hbar\omega\rangle$  and the  $|x, (n-1)\hbar\omega\rangle$  lies above the  $|g, n\hbar\omega\rangle$  for  $\Omega \ge E_b$  (Figure 3b,c) the repulsion between the Floquet and the original states cause the exciton resonance to blueshift by  $\delta_x$ . The illustration of the coherent signal resulting from blueshift of exciton resonance is shown in Figure 3f. There is no contribution from the biexciton interaction in  $\sigma^+\sigma^+$ , the theoretical description is exactly the same as that of the ordinary OSE in a non-interacting framework  $^{10,12,58}$ .

To make a quantitative comparison between experiment and theory in the interacting regime, we numerically simulated the spectrum at different  $\Omega$  by solving the effective Hamiltonian in Equation 1 and Supporting Information. The simulation is accomplished by using homogeneous broadening of 100 meV and biexciton binding energy of 71 meV. The Rabi energy extracted is  $V_{xx} \approx V_x = 83 \pm 2$  meV and corresponding  $\mu_{xx} \approx \mu_x = 25 \pm 1$  Debye. The biexciton transition dipole moment is larger than those of any known systems, such as CuCl (~20 Debye), monolayer MoSe<sub>2</sub> (9.3 Debye), and GaAs/AlGaAs quantum dot (10 Debye)<sup>17,24,29</sup>. Figure 4a shows that calculated  $\Delta A$  spectrum of  $\sigma^+\sigma^-$  for a pump fluence of 1 mJ/cm<sup>2</sup>, presented as pseudo-color plot, at different detuning energies reasonably matches with the experimental  $\Delta A$  spectrum. Moreover, the blueshift and redshift for the  $\sigma^+\sigma^-$  and blueshift in  $\sigma^+\sigma^+$ , is exactly matching with our theoretical calculations, solid lines in Figure 2c.

The other prominent signature of strong coherent exciton-biexciton interactions is obtained from the fluence-dependent  $\Delta A$  for  $\sigma^+\sigma^-$  and  $\sigma^+\sigma^+$ . Similar to detuning energy dependence, the

Jaynes-Cummings model can be employed to account for the fluence dependence as well. For instance, the pseudo-color plots in Figure 4b demonstrates the excellent agreement between experimentally and theoretically calculated  $\Delta A$  spectra ( $\sigma^+\sigma^-$ ) at different pump-fluences for  $\Omega$  = 82 meV.

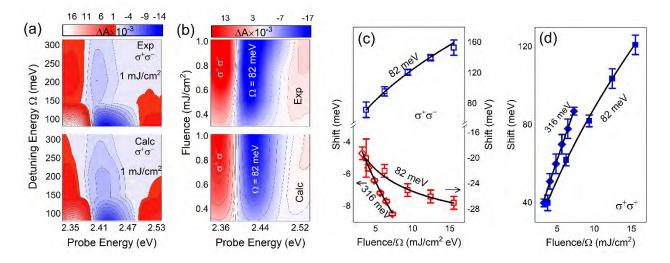


Figure 4. Detuning and fluence dependent OSE. Pseudo-colour plot of the experimental (top) and theoretically calculated (bottom)  $\Delta A$  at zero pump-probe delay as a function of (a)  $\Omega$  at constant fluence of  $1 \text{mJ/cm}^2$  and of (b) fluence at constant  $\Omega = 82 \text{ meV}$ , under  $\sigma^+ \sigma^-$ ; The optical stark shift evaluated experimentally (symbols) and theoretically calculated (solid lines) at different fluences at  $\Omega = 316 \text{ meV}$  and 82 meV for (c)  $\sigma^+ \sigma^-$  and (d)  $\sigma^+ \sigma^+$ .

The optical Stark shift evaluated at different fluences at two extreme  $\Omega$  used in present study, 316 and 82 meV, is shown in Figure 4c for  $\sigma^+\sigma^-$  and Figure 4d for  $\sigma^+\sigma^+$ . Under  $\Omega=316$  meV, the optical stark shift for  $\sigma^+\sigma^-$  (hollow red diamond) and  $\sigma^+\sigma^+$  (solid blue diamond) show a linear fluence dependence, which can be retrieved by including large detuning approximation ( $\Omega>>> V_x$ ) to Equation 2 and 4, that reduces to

$$\delta_{xx_R} = \frac{v_{xx}^2}{|\Omega| - E_h} - \frac{v_x^2}{|\Omega|} \quad and \quad \delta_{x_B} = \frac{2v_x^2}{|\Omega|}$$
 (5)

This picture changes dramatically for  $\Omega = 82$  meV. Here, the optical Stark redshift (hollow red

square) and blueshift (hollow blue square) for  $\sigma^+\sigma^-$  and blueshift (solid blue square) for  $\sigma^+\sigma^+$ 

exhibit a non-linear fluence dependence in accordance with Equation 2-4, as shown in Figure 4c,d,

in stark contrast to the previous reports in two-dimensional<sup>12</sup> and bulk perovskites<sup>11</sup>.

In summary, we have demonstrated the first observation of anomalous excitonic OSE at

room temperature in solution processed CsPbBr<sub>3</sub> QDs that necessitate the consideration of the

biexciton interactions for a complete description. Further, we anticipate that these types of coherent

biexciton states can offer a pathway to explore solution processed QDs based room temperature

quantum information processing, lasing, mode-locking, electromagnetically induced transparency,

and to generate a pulse of light containing many photons in a highly entangled state for which the

polarization state of each photon is intricately linked to others – a new resource for distributed

quantum technologies.

ASSOCIATED CONTENT

Supporting Information. Synthesis of CsPbBr<sub>3</sub> QDs, circularly polarized transient absorption

spectroscopy, determination of exciton binding energy through Elliott model and temperature

dependent PL, Biexciton binding energy determination through fluence dependent PL and transient

absorption measurements, detuning dependent OSE, time evolution of features, and theoretical

description in interacting framework.

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**Notes** 

The authors declare no competing financial interest.

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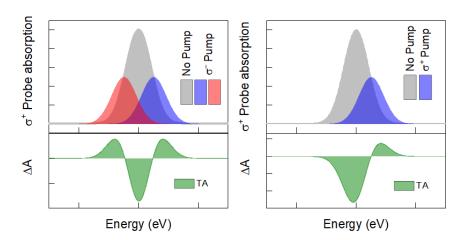
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# TABLE OF CONTENT GRAPHIC



# **Correction:**

# **Room-Temperature Anomalous Coherent Excitonic Optical Stark**

### **Effect in Metal Halide Perovskite Quantum Dots**

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The panel (a) and (b) of Figure 2 of this article were inadvertently swapped during production of final files. The correct version of the figure appears below.

