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Attractive and repulsive dipolar interaction in bilayers of indirect excitons

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We explore attractive dipolar interaction in indirect excitons (IXs). For one layer of IXs in a single pair of coupled quantum wells (CQW), the out-of-plane IX electric dipoles lead to repulsive dipolar interaction between IXs. The attractive dipolar interaction between IXs is realized in a 2-CQW heterostructure with two IX layers in two separated CQW pairs. We found both in experimental measurements and theoretical simulations that increasing density of IXs in one layer causes a monotonic energy reduction for IXs in the other layer. We also found an in-plane shift of a cloud of IXs in one layer towards a cloud of IXs in the other layer. This behaviour is qualitatively consistent with attractive dipolar interaction. The measured IX energy reduction and IX cloud shift are higher than the values given by the correlated liquid theory.

PACS numbers:

I. INTRODUCTION

A spatially indirect exciton (IX), also known as an interlayer exciton, is a bound pair of an electron and a hole confined in separated layers. Due to the electron-hole separation, IXs have built-in electric dipole moment ed , where d is the distance between the electron and hole layers and e electron charge. Furthermore, due to the electron-hole separation, IXs have long lifetimes within which they can cool below the temperature of quantum degeneracy [1]. These properties make IXs a platform for exploring quantum gases with dipolar interaction.

IXs can be realized in a pair of quantum wells separated by a narrow tunneling barrier. For one layer of IXs in a single pair of coupled quantum wells (CQW), the out-of-plane IX electric dipoles lead to repulsive dipolar interaction between side-to-side IX dipoles (Fig. 1). This configuration is extensively studied both theoretically and experimentally. The phenomena originating from the repulsive dipolar interaction in a single IX layer include the enhancement of IX energy with density that has been known since early studies of IXs [2–6], screening of in-plane disorder potential by repulsively interacting IXs [7–10] that leads to IX delocalization and long-range IX transport [7–18], strong correlations [10, 19–26], and predicted crystal phases [27–32].

Quantum gases with dipolar interaction are also explored with cold atoms. In these systems, dipolar interactions lead to droplet structures with spatial ordering and coherence [36–40], few-body complexes [41, 42], and pair superfluid and crystal phases in bilayers of dipoles [43–46].

The specific property of dipolar interaction is its anisotropy. For instance, for two parallel dipoles tilted at angle θ relative to the line connecting them, the interaction at $r \gg d_1, d_2$ is given by $v(r) \sim e^2 d_1 d_2 (1 - 3 \cos^2 \theta) / \epsilon r^3$, where ϵ is the dielectric constant of the material and $p_{1,2} = ed_{1,2}$ the dipole moments [47]. For the out-of-plane

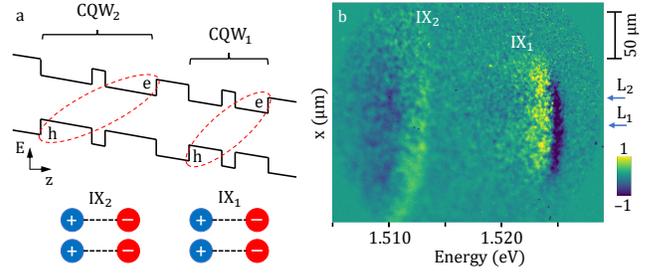


FIG. 1: **2-CQW heterostructure and x -energy image.** (a) Diagram of 2-CQW structure with two CQW pairs. IX_2 form in CQW₂, IX_1 in CQW₁. Schematic below shows IX dipoles. – electrons, + holes. The intra-CQW interaction between IX_2 (or IX_1) side-to-side dipoles is repulsive. The inter-CQW interaction between IX_2 and IX_1 head-to-tail dipoles is attractive. (b) Differential x -energy luminescence image. The arrows indicate the excitation spot positions of L_2 and L_1 lasers resonant to direct excitons in 15 nm CQW₂ and 12 nm CQW₁, respectively. L_2 generates IX_2 . L_1 generates IX_1 and also a smaller concentration of IX_2 . The laser powers $P_{L1} = 10 \mu W$, $P_{L2} = 250 \mu W$. The differential x -energy image is obtained by subtracting the x -energy images created by only L_1 on and by only L_2 on from the x -energy image created by both lasers on. In the differential x -energy image, the yellow and blue colors indicate the enhancement and reduction of the IX luminescence intensity, respectively. For the energy axis, blue region on the right side and yellow on the left (observed for IX_1) corresponds to an energy decrease. For the position axis, blue region on the low side and yellow on the high (observed for IX_1) corresponds to a cloud shift up. The differential x -energy image shows an increase of IX_2 energy, a decrease of IX_1 energy, and a spatial shift of the IX_1 cloud towards the IX_2 cloud.

IX dipoles in a single IX layer this expression reduces to $v(r) \sim e^2 d^2 / \epsilon r^3$ describing the repulsive dipolar interaction between IXs.

The other specific property of dipolar interaction for the IXs is the induced orientation of IX dipoles. The heterostructure design and/or applied voltage, which pro-

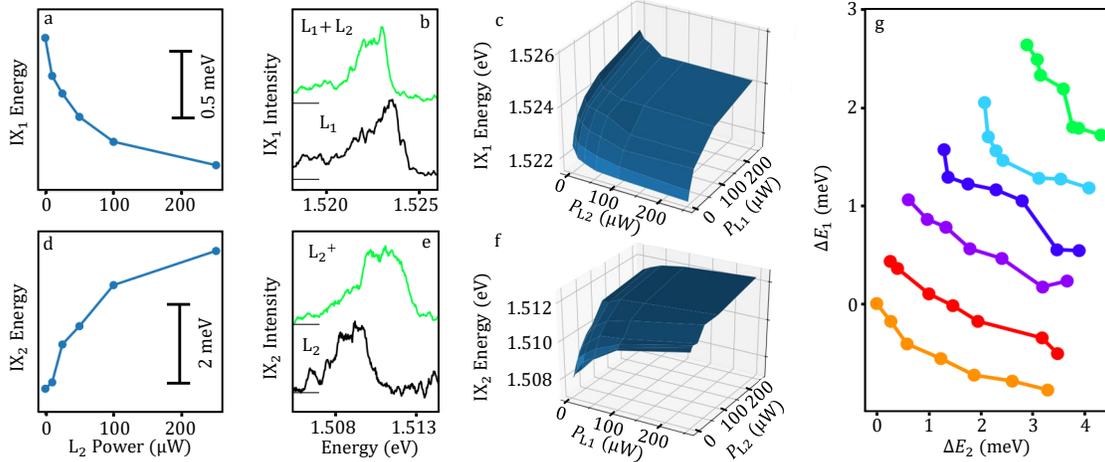


FIG. 2: **Decrease and increase of IX energy due to attractive and repulsive dipolar IX interaction: Experiment.** (a) The decrease of IX_1 energy with increasing L_2 power and, in turn, IX_2 density. $P_{L1} = 10 \mu W$. An energy decrease corresponds to attractive $IX_1 - IX_2$ interaction. (b) The red shift of IX_1 spectrum with turning on L_2 , which increases IX_2 density. Black line shows IX_1 spectrum when only L_1 is on, $P_{L1} = 10 \mu W$. Green line shows IX_1 spectrum when additional L_2 is on, $P_{L2} = 250 \mu W$. (c) IX_1 energy as a function of both P_{L1} and P_{L2} . (d) The increase of IX_2 energy with increasing L_2 power and, in turn, IX_2 density. $P_{L1} = 0$. An energy increase corresponds to repulsive $IX_2 - IX_2$ interaction. (e) The blue shift of IX_2 spectrum with increasing L_2 power, which increases IX_2 density. $P_{L2} = 25 \mu W$ (black line) and $250 \mu W$ (green line). (f) IX_2 energy as a function of both P_{L1} and P_{L2} . (g) The change in IX_1 energy vs the change of IX_2 energy. Each set of data corresponds to increasing L_2 power. For the sets of data presented by orange, red, purple, blue, cyan, and green points, $L_1 = 5, 10, 25, 50, 100,$ and $250 \mu W$, respectively.

duces the electric field in the heterostructure, determine the quantum well layers where electrons and holes are confined: Exchanging the quantum wells by the electron and the hole, i.e. flipping the IX dipole, is energetically unfavorable. Furthermore, tilting the IX dipole relative to the z direction causes an in-plane separation of the electron and the hole in the IX and, as a result, reduces the IX binding energy. This induces the orientation of IX dipoles in the direction normal to the QW plane.

The induced orientation of the IX dipoles and the repulsive dipolar interaction for a single IX layer makes it challenging to explore the attractive dipolar interaction in IX systems. The studies of IX dipoles have been concentrated on the case of repulsively interacting IXs [1–31]. However, the angle-dependent dipolar IX interaction and, in particular, dipolar attraction gives an access to new phenomena in quantum dipolar gases. For instance, the dipolar attraction leads to the phenomena in cold atoms outlined above [36–46]. IX attraction can be realized by extending IX heterostructures beyond a single CQW design and studies of attractively interacting IX dipoles were recently started in two stacked CQW pairs [48–50].

In this work, we explore the attractive dipolar interaction between IXs in a 2-CQW heterostructure with two IX layers in two separated CQW pairs (Fig. 1). The intra-CQW interaction between IX side-to-side dipoles is repulsive similar to single CQW heterostructures [1–

31]. The inter-CQW interaction between IX head-to-tail dipoles is attractive. It changes to repulsive with increasing in-plane separation between the IXs and, in turn, θ following the anisotropy of dipolar interaction outlined above. Both our experimental measurements and theoretical simulations show (i) a monotonic energy reduction for IXs in one layer with increasing density of IXs in the other layer and (ii) an in-plane shift of a cloud of IXs in one layer towards a cloud of IXs in the other layer. This behaviour is qualitatively consistent with attractive dipolar interaction, however, the measured IX energy reduction and IX cloud shift are higher than the values given by the correlated liquid theory.

II. EXPERIMENT

The studied 2-CQW heterostructure (Fig. 1a) is grown by molecular beam epitaxy. Indirect excitons IX_2 form in CQW_2 , indirect excitons IX_1 in CQW_1 . CQW_2 consist of two 15 nm GaAs QWs separated by 4 nm $Al_{0.33}Ga_{0.67}As$ barrier, CQW_1 consists of two 12 nm GaAs QWs separated by 4 nm $Al_{0.33}Ga_{0.67}As$ barrier. CQW_2 and CQW_1 are separated by 12 nm $Al_{0.33}Ga_{0.67}As$ barrier, narrow enough to allow substantial inter-layer interaction between IX_2 and IX_1 , yet wide enough to suppress tunneling of electrons and holes between CQW_2 and CQW_1 . n^+ -GaAs layer with $n_{Si} \sim 10^{18} \text{ cm}^{-3}$ serves as a bottom

electrode. The CQW pair is positioned 100 nm above the n^+ -GaAs layer within undoped 1 μm thick $\text{Al}_{0.33}\text{Ga}_{0.67}\text{As}$ layer. The two CQW pairs are positioned closer to the homogeneous bottom electrode to suppress the fringing in-plane electric field in excitonic devices [51]. The top semitransparent electrode is fabricated by applying 2 nm Ti and 7 nm Pt on 7.5 nm GaAs cap layer. Applied gate voltage $V_g = -2$ V creates electric field in the z -direction. The cross-section of the heterostructure is presented in Appendix D.

The IX_2 energy is lower than the IX_1 energy. This energy difference gives an opportunity to selectively generate IX_2 by optical excitation. Excitons are generated by semiconductor lasers L_2 and L_1 at the energies 1.532 and 1.541 eV resonant to spatially direct excitons (DXs) in CQW_2 and CQW_1 , respectively. The resonant to DX excitation increases the light absorption and, in turn, IX density for a given laser power [52]. L_2 generates IX_2 . L_1 generates IX_1 and also roughly 2 times smaller concentration of IX_2 due to a weaker nonresonant absorption of L_1 light in CQW_2 . L_2 and L_1 excitations are focused to ~ 5 μm hwhm spots, which are separated by 50 μm . This configuration allows exploring the effects of IX interactions on the IX cloud position. IX photoluminescence (PL) is measured in a 20 ns time window starting 20 ns after the end of the L_1 and/or L_2 excitation pulses. This allows for studying of only long-lived IX s after DXs recombined. Both IX_2 and IX_1 have long lifetime in the range of hundreds of ns (~ 800 ns for IX_2 and ~ 260 ns for IX_1) allowing them to travel over long distances reaching hundreds of microns.

Time-resolved imaging experiments are performed with a laser pulse duration 2000 ns, period 4000 ns, and edge sharpness ~ 2 ns. The rectangular-shaped pulses are realized by a pulse generator driving the semiconductor lasers. The pulse duration and period are optimized to allow the IX PL image to approach equilibrium during the laser excitation and decay between laser pulses. The PL images are captured using a PicoStar HR TauTec time-gated intensifier. The PL passes through a spectrometer with a resolution of 0.18 meV before entering the intensifier coupled to a liquid-nitrogen-cooled CCD. The measurements are performed at $T_{\text{bath}} = 1.7$ K.

To analyze the attractive inter-layer IX interaction in the IX bilayer we measure how the selective generation of IX_2 affects the energies and cloud position of IX_1 . Figure 1b presents the differential x -energy image obtained by subtracting the x -energy images created by only L_1 on (Fig. 10c in Appendix C) and by only L_2 on (Fig. 10b) from the x -energy image created by both lasers on simultaneously (Fig. 10a). The differential x -energy image shows an increase of IX_2 energy, a decrease of IX_1 energy, and a spatial shift of the IX_1 cloud towards the IX_2 cloud. These phenomena are detailed below.

First, we consider the IX energy variations. Figures 2a-c show that the increase of L_2 power (P_{L2}) and, in turn,

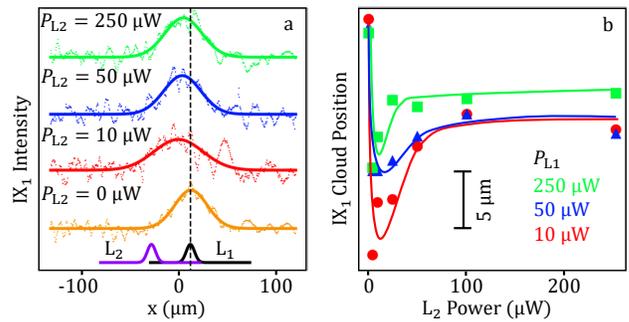


FIG. 3: **Attraction of the IX_1 cloud to the IX_2 cloud: Experiment.** (a) The IX_1 cloud profiles at different P_{L2} and, in turn, IX_2 densities. $P_{L1} = 10$ μW . The profiles of L_1 and L_2 laser excitation spots are shown by black and purple lines, respectively. Dashed line indicates the center of L_1 excitation spot. (b) The center of mass position of the IX_1 cloud as a function of P_{L2} for different P_{L1} .

IX_2 density (n_2) leads to a monotonic decrease of IX_1 energy. An energy decrease corresponds to attractive $\text{IX}_1 - \text{IX}_2$ interaction. In comparison, when only IX_2 are present in the system (L_1 is off), the increase of P_{L2} and, in turn, n_2 leads to a monotonic increase of IX_2 energy (Fig. 2d-f). An energy increase corresponds to repulsive $\text{IX}_2 - \text{IX}_2$ interaction, which has been extensively studied in single layers of IX s [1–31].

Figures 2c,f also show that the increase of P_{L1} leads to a monotonic increase of both IX_1 and IX_2 energies. L_1 generates both IX_1 and IX_2 , therefore, the effect of increasing P_{L1} on IX_1 (or IX_2) energy is a combined effect of attractive $\text{IX}_1 - \text{IX}_2$ and repulsive $\text{IX}_1 - \text{IX}_1$ (or $\text{IX}_2 - \text{IX}_2$) interactions. The monotonic increase of both IX_1 and IX_2 energies with P_{L1} indicates that the repulsive interaction is stronger. This is consistent with the relative strength of the attractive (Fig. 2a) and repulsive (Fig. 2d) interaction in the experiments with increasing P_{L2} which increase only IX_2 density.

In the mean-field approximation the repulsive interaction between IX s in a single layer increases the IX energy by $\Delta E = 4\pi e^2 dn/\epsilon$. This equation known as the “plate capacitor” formula provides a qualitative explanation for the observed monotonic increase of ΔE with the exciton density n [2]. However, the capacitor formula can significantly overestimate $\Delta E(n)$ due to the IX correlations [10, 19–26]. To compare the attractive and repulsive dipolar interactions, avoiding the complexity of the relation between ΔE and n , Fig. 2g presents the change in IX_1 energy ΔE_1 vs the change of IX_2 energy ΔE_2 for the data in Fig. 2c,f. The energy shifts ΔE are measured relative to the IX energies at the lowest n . Figure 2g shows that for all studied P_{L1} , the increase of P_{L2} and, in turn, n_2 leads to ΔE_2 larger in absolute value than ΔE_1 , indicating that the repulsive $\text{IX}_2 - \text{IX}_2$ interaction is stronger than the attractive $\text{IX}_2 - \text{IX}_1$ interaction.

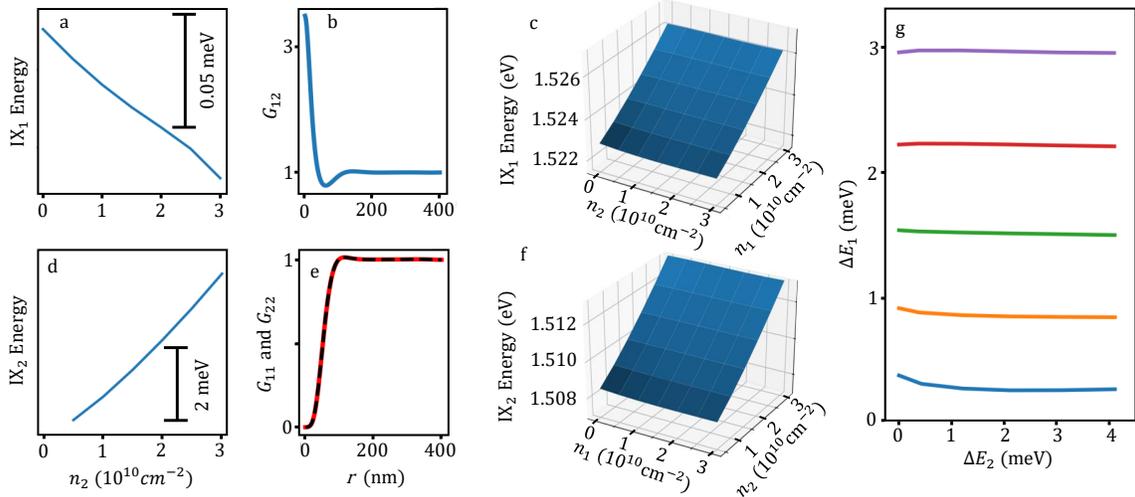


FIG. 4: **Decrease and increase of IX energy due to attractive and repulsive dipolar IX interaction: Theory.** (a) The decrease of IX₁ energy with increasing IX₂ density. $n_1 = 1.5 \times 10^{10} \text{ cm}^{-2}$. An energy decrease corresponds to attractive IX₂ – IX₁ interaction. (b) IX₁ – IX₂ density correlation function. $n_1 = n_2 = 10^{10} \text{ cm}^{-2}$. (c) IX₁ energy as a function of both IX₁ and IX₂ density. (d) The increase of IX₂ energy with increasing IX₂ density. $n_1 = 0$. An energy increase corresponds to repulsive IX₂ – IX₂ interaction. (e) IX₂ – IX₂ (red solid line) and IX₁ – IX₁ (black dashed line) density correlation functions. $n_1 = n_2 = 10^{10} \text{ cm}^{-2}$. (f) IX₂ energy as a function of both IX₁ and IX₂ density. (g) The change in IX₁ energy vs the change of IX₂ energy. Each line corresponds to increasing IX₂ density. For the blue, orange, green, red, and purple lines, $n_1 = 0.5, 1, 1.5, 2, \text{ and } 2.5 \times 10^{10} \text{ cm}^{-2}$, respectively.

We also consider the spatial shift of the IX₁ cloud toward the IX₂ cloud [53]. Figure 3 shows that the IX₁ cloud attracts to the IX₂ cloud. With increasing P_{L2} , the spatial shift is nonmonotonic. This behaviour is observed for different P_{L1} . A larger spatial shift, reaching $\sim 10 \mu\text{m}$, is observed at low P_{L1} (Fig. 3b).

III. THEORY

We analyze the dipolar interaction in IX bilayers theoretically and compare the experimental data with theoretical simulations. The numerical simulation of such two-species many-body system is done through the Hyper-netted Chain (HNC) formalism [54]. The intra- and inter-layer interaction is modelled by assuming that the wave-function of electrons and holes are isotropic Gaussians and they experience a Coulomb interaction, see Appendix A. HNC method has been previously used for studying bosons with dipolar interactions in single-layer systems [55]. Unless the interaction strength is very high, the HNC predictions for basic many-body properties such as pair-correlation functions and energy per particle were shown to be in a good agreement with the more accurate Monte-Carlo calculations [29, 45, 46].

The simulated IX energy shifts caused by the attractive and repulsive IX dipolar interactions are presented in Fig. 4. Figures 4a,c show that the increase of n_2 leads to a monotonic decrease of IX₁ energy due to the attractive

IX₁ – IX₂ interaction, in qualitative agreement with the experimental data in Fig. 2a,c. In contrast, the increase of n_2 (or n_1) leads to a monotonic increase of IX₂ (or IX₁) energy (Fig. 4d,f) due to the repulsive IX₂ – IX₂ (or IX₁ – IX₁) interaction, in qualitative agreement with the experimental data in Fig. 2d,f.

In the simulations, n_1 is increased selectively keeping n_2 intact. In the experiment, an increase of n_1 is accompanied by an increase of n_2 as outlined above. This leads to different variations of IX₂ energy with n_1 in the experiment (Fig. 2f) and the theory (Fig. 4f). However, the conclusions on the attractive IX₁ – IX₂ interaction and repulsive IX₂ – IX₂ interaction derived from the experiment and the comparison between the experiment and the theory are based on the n_2 dependence and are not affected by the difference in the n_1 dependence.

The density correlation functions for the cases of attractive IX₁ – IX₂ and repulsive IX₁ – IX₁ and IX₂ – IX₂ interactions are presented in Fig. 4b and 4e, respectively. For the attractively interacting IXs (Fig. 4b), the correlation function enhancement above 1, the mean-field value, increases the interaction energy compared with the vanishing interlayer interaction in the mean-field approximation. On the contrary, the repulsively interacting IXs avoid each other (Fig. 4e) that lowers the intralayer IX interaction energy compared with the uncorrelated state assumed in the mean-field approximation.

The energy shift in Fig. 4d is close to 1/3 of the shift given by the “plate capacitor” formula, in other words,

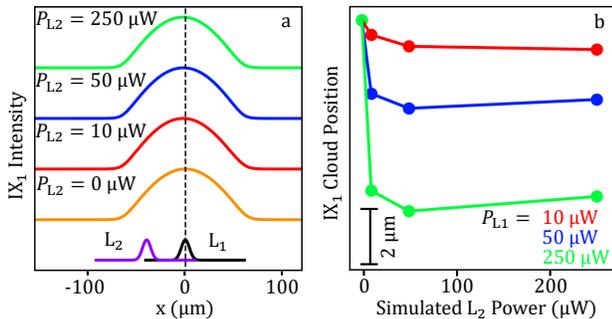


FIG. 5: **Attraction of the IX₁ cloud to the IX₂ cloud: Theory.** (a) The IX₁ cloud profiles simulated for different P_{L2} and, in turn, IX₂ densities. P_{L1} = 250 μW. The profiles of L₁ and L₂ laser excitation spots are shown by black and purple lines, respectively. Dashed line indicates the center of L₁ excitation spot. (b) The center of mass position of the IX₁ cloud as a function of P_{L2} for different P_{L1}.

the exciton density is related to the measured energy shift caused by the intra-layer IX interaction by the formula $\Delta E \sim 1/3 \cdot 4\pi e^2 dn/\epsilon$. In Figs. 2d and 4d, the ranges of the energy shift in the experiment and the theory are similar, so that the range of the L₂ laser power in Fig. 2d approximately corresponds to the range of the exciton density n_2 in Fig. 4d.

As for the experimental data, we compare the attractive and repulsive dipolar interactions in a graph showing ΔE_1 vs ΔE_2 . Figure 4g shows that for all studied n_1 , the increase of n_2 leads to calculated ΔE_2 larger in absolute value than ΔE_1 , in qualitative agreement with the experimental data in Fig. 2g.

We also simulated the spatial shifts of the IX₁ cloud toward the IX₂ cloud. The simulations of the IX spatial profiles are based on the IX generation, diffusion, and recombination and are outlined in Appendix B. Figure 5 shows that the IX₁ cloud attracts to the IX₂ cloud in the simulations, in qualitative agreement with the attraction observed in the experiment (Fig. 3). In comparison, both our experimental measurements (Fig. 8) and theoretical simulations (Fig. 9) show that two clouds of repulsively interacting IX₂ repel each other, see Appendix B.

While both the experimental measurements and theoretical simulation show (i) a monotonic IX₁ energy reduction with increasing IX₂ density and (ii) an in-plane shift of IX₁ cloud towards IX₂ cloud, consistent with attractive dipolar interaction, the measured IX₁ energy reduction and IX₁ cloud shift are higher than the values given by the correlated liquid theory (compare Figs. 2a and 4a, Figs. 2g and 4g, and Figs. 3 and 5). The interaction-and/or disorder-induced IX mass enhancement may be one possible reason for this discrepancy. The magnitude of IX₁ energy reduction, ΔE_1 , scales with the strength of interlayer IX₁ – IX₂ dipolar attraction (Fig. 6). For the

case of a single IX₁ – IX₂ pair, ΔE_1 can be estimated as the binding energy of IX₁ – IX₂ biexciton state, E_b . For a bare IX mass, $m_{IX} \sim 0.2m_0$ [56], $E_b \sim 0.3$ meV (Fig. 7). Higher E_b can be achieved for higher IX masses, and, e.g. for the reduced IX mass enhanced to $2m_0$, E_b reaches ~ 1.2 meV (Fig. 7), making the IX₁ – IX₂ interaction scale comparable to the experiment (Fig. 2). A mass enhancement can be caused by interaction, however, only a relatively weak interaction-induced mass enhancement, up to $\sim 25\%$, was observed in electron-hole systems in single QWs [57]. The studies of effects of interaction and/or disorder on the IX mass can be a subject of future work.

Layered heterostructures with more than one IX layer, like IX bilayers in this work, and, generally, with more than two electron and hole layers, like three-layer heterostructures in Ref. [58], could be new platforms for studying systems with attractive dipolar interaction. For heterostructures with a fixed set of materials, the range of parameter variation may be limited. For instance, for the GaAs/AlGaAs heterostructures used in this work, it is difficult to achieve a significant change of the dipolar attraction between the IXs by varying the AlGaAs spacer layer thickness d_s : A significant reduction of d_s would lead to substantial tunneling of electrons and holes between CQW₁ and CQW₂, destroying both IX₁ and IX₂, while a significant enhancement of d_s would lead to a substantial reduction of the dipolar interaction, which drops as $1/r^3$ and is already weak for $d_s = 12$ nm in the heterostructure studied in this work.

However, a significant change of the parameters can be achieved by using different materials. In particular, van der Waals heterostructures offer an opportunity to study IX bilayers with small d and/or d_s (significantly smaller than in the GaAs heterostructures) and high IX binding energies (significantly higher than in the GaAs heterostructures), see Ref. [59] and references therein. The studies of dipolar interactions in IX bilayers and other IX systems with multiple electron and hole layers in van der Waals heterostructures can be a subject of future work.

IV. CONCLUSION

In summary, we presented experimental and theoretical studies of attractive dipolar interaction in IX bilayers. We found that increasing density of IXs in one layer causes a monotonic energy reduction for IXs in the other layer. We also found an in-plane shift of a cloud of IXs in one layer towards a cloud of IXs in the other layer. This behaviour is qualitatively consistent with attractive dipolar interaction. The measured IX energy reduction and IX cloud shift are higher than the values given by the correlated liquid theory.

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APPENDIX A: EXCITON INTERACTION

Interaction potentials

We modeled IXs as composite bosonic particles with a rigid internal charge distribution. The interactions of such particles can be specified in terms of potentials $u_{ij}(r)$, $1 \leq i, j \leq 2$, which are functions of pairwise in-plane distances r of the excitons. Here and below we use subscripts 1 and 2 to label the CQWs (12 nm- and 15 nm-wide, respectively). To compute these interaction potentials, we assumed that the charge distributions of all the electrons and holes are spherically symmetric Gaussians. The radius a of the Gaussians is our adjustable parameter that accounts for the width of the quantum wells and the internal motion of particles about the center of mass of each exciton. We computed these interaction potentials by taking the convolutions of the Coulomb kernel $e^2/\epsilon\sqrt{r^2+z^2}$ with the charge densities of the interacting particle pairs. The result for the inter-CQW potential $u_{12}(r) = u_{21}(r)$ is

$$u_{12}(r) = \sum_{\sigma=\pm} \sum_{\tau=\pm} V\left(\sqrt{r^2+z_{\sigma\tau}^2}\right), \quad (1)$$

$$V(r) = \frac{e^2}{\epsilon r} \operatorname{erf}\left(\frac{r}{2a}\right), \quad (2)$$

$$z_{\sigma\tau} = \tau \frac{d_1 - \sigma d_2}{2} + D, \quad (3)$$

where $\operatorname{erf}(x)$ is the error function and D is the z-axis distance between the CQW centers. The intra-CQW potentials $u_{kk}(r)$, $k = 1$ or 2 , are given by the same equation with $D = 0$ and the electron-hole separations d_1, d_2 replaced by d_k . The plots of these potentials for parameter values representative of our experimental device are shown in Fig. 6. At $r \ll a$ all of them approach constant finite values and at large r , these potentials behave as $1/r^3$. Potentials $u_{11}(r)$ and $u_{22}(r)$ are strictly repulsive. Potential $u_{12}(r)$ is attractive in the range of distances r selected for the plot. At larger r , it eventually becomes repulsive but it is already very small as such r .

The potentials $u_{ij}(r)$ serve as inputs to our computer program that computes many-body properties within the zero-temperature hypernetted chain (HNC) formalism. Another input parameter is the effective mass m_{IX} of the excitons, which we took to be 0.2 of the free electron mass m_0 . Our implementation of the HNC method is based on Ref. [54]. The output of these calculations include the pair correlation functions $G_{ij}(r)$, the energy

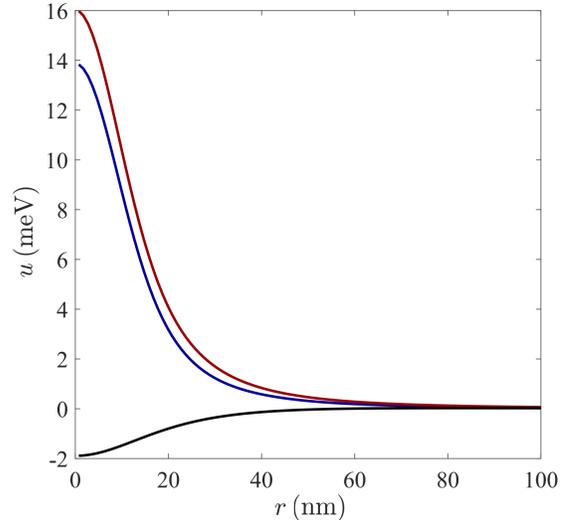


FIG. 6: Model interaction potentials: u_{22} (top) and u_{11} (middle) are the intra-CQW potentials for IX₂ and IX₁, respectively; u_{12} (bottom curve) is the inter-CQW interaction potential. Parameters: $d_1 = 20, d_2 = 25, D = 43, a = 5$ (all in nm); $\epsilon = 13$.

density $\epsilon = \epsilon(n_1, n_2)$, and the chemical potentials

$$\mu_j = \partial\epsilon/\partial n_j \quad (4)$$

of the excitons as functions of their number densities n_1 and n_2 in the CQWs. If the shake-up effects, i.e., many-body relaxation processes following the exciton recombination, can be neglected, then the exciton emission energies (or exciton “single-particle energies”) E_j should coincide with their chemical potentials:

$$E_j \approx \mu_j. \quad (5)$$

Based on this assumption, we have constructed the plot of ΔE_1 vs. ΔE_2 shown in Fig. 4g of the main text. Representative intra-CQW and inter-CQW pair-correlation functions are plotted in Fig. 4b,e. At short distances, these functions show a deep “correlation hole” for excitons of the same CQW and a strong correlation peak for excitons of different CQWs.

Biexciton binding energy

From previous theoretical work on double-layer bosonic systems with repulsive intra-layer and attractive inter-layer dipolar interactions [43–46], we expect that the exciton system at low enough equal densities $n_1 = n_2$ should be made of bound pairs, the inter-CQW biexcitons. If $n_1 \neq n_2$, then biexcitons and unpaired excitons may co-exist. A rough estimate of the required density is given by the Mott criterion stating that the

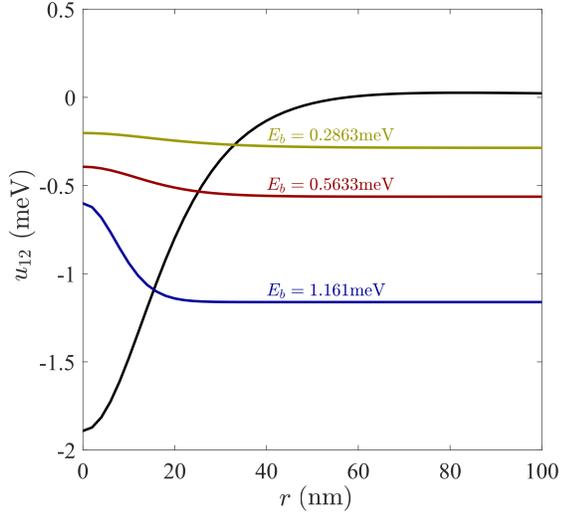


FIG. 7: Probability density distributions of inter-CQW biexcitons within the rigid-body approximation. The interaction potential u_{12} (same as in Fig. 6) is shown by the curve with a dip (black curve). The probability distributions (wavefunctions squared) of the bound states are depicted by the curves with the maxima. These curves are plotted in arbitrary units and are offset to show the binding energies E_b . They correspond (top to bottom) to the reduced masses of 0.5, 1.0, and 10 of the exciton mass.

biexcitons should appear when the dimensionless parameter $\min(n_1, n_2) b^2$ is less than some critical number, which is usually numerically small, perhaps, 0.02. Here b is the spatial size of the biexciton. Within our rigid-body approximation, the biexciton bound state can be easily found numerically. In the relative coordinates, this problem reduces to solving a Schrödinger equation for a particle of reduced mass $m_{IX}/2$ subject to the confining potential $u_{12}(r)$. For the same parameters as in Fig. 6, we obtained the binding energy to be $E_b = 0.286$ meV. From Fig. 7 we deduce the spatial size of the biexciton to be $b \sim 30$ nm, so that the Mott critical density for biexcitons is of the order of 10^{10} cm $^{-2}$, not too far from the exciton densities realized in our experiment.

The following argument suggests that E_b is in fact the maximum possible shift of the single-particle energies due to the inter-CQW attraction. Indeed, in the limit of high densities, where average intra-CQW exciton separation is smaller than D , correlations are negligible. At intermediate densities, where HNC should be accurate, the dependence of say $E_1 = E_1(n_1, n_2)$ on n_2 with n_1 held fixed is either monotonic or flat within computational accuracy, see Fig. 4c,f. Therefore, the asymptotic limit $n_1 = n_2 \rightarrow 0$, where all excitons are paired and

$$E_1 = E_2 = -E_b \quad (6)$$

should correspond to the largest possible attraction ef-

fect. Note that the HNC method reproduces this asymptotic limit only approximately. The tendency toward pairing is manifested in the aforementioned peak in the pair-correlation function $G_{12}(r)$ at $r = 0$. The shape of this peak computed by the HNC resembles the probability distribution of the biexciton, cf. Figs. 4b and 7. The integrated weight $N = n_2 \int G_{12}(r) d^2r$ of the peak (where the integration extends up to $r \sim b$) is the average number of excitons of CQW $_1$ attracted to an exciton in CQW $_2$. When the biexcitons form, N should approach unity. Yet within our HNC calculations N keeps increasing as $n_1 = n_2$ decreases. This suggests that the standard HNC method is inadequate in the low-density regime where we should instead use Eq. (6).

We found both in experimental measurements (Fig. 2a,c) and theoretical simulations (Fig. 4a,c) that increasing density of IXs in one layer causes a monotonic energy reduction for IXs in the other layer. These results differ with the results of Refs. [49, 50] where a nonmonotonic dependence on the density was reported. The nonmonotonic dependence on the density was attributed to many-body polaron effects in Refs. [49, 50]. Our simulations show no indication for the non-monotonic dependence on the density.

The experiment still poses a challenge for the theory because the shift of E_1 has been observed to routinely exceed the computed $E_b = 0.286$ meV, see Fig. 2a. To identify a possible reason for the discrepancy, we examined this important parameter more critically. First, we tested the validity of the rigid-body approximation. We used a previously developed computational tool [34] to accurately solve for the exciton and biexciton binding energies as two-body and four-body problems, respectively. For the parameters of Fig. 6 we obtained $E_b = 0.33$ meV. Hence, the rigid-body approximation is not the major source of the discrepancy. Next, we noticed that E_b is greatly reduced compared to the depth ≈ 2 meV of the potential well $u_{12}(r)$. This reduction is due to the zero-point motion. As an illustration of how this quantum effect may affect the binding energy, we recalculated E_b and the wavefunctions of biexcitons for reduced masses enhanced two- and twenty-fold. In the latter case, the binding energy rises to 1.16 meV, see Fig. 7, which is close to the experimentally measured shifts of E_1 we attributed to the inter-CQW attraction. It is hard to expect that the exciton mass is indeed enhanced by such an enormous factor due to the interaction alone. (As a point of reference, only a relatively weak interaction-induced mass enhancement, up to 25%, was observed in electron-hole systems in single QWs [57].) However, the suppression of the zero-point motion of an exciton pair may in principle be facilitated by disorder in the system that traps the excitons close together in deep potential wells.

APPENDIX B: DYNAMICS OF EXCITON DENSITY DISTRIBUTION

In this section we summarize the set of equations we used to model the macroscopic dynamics of excitons. To simplify the modeling, we assumed that the exciton densities n_k and currents j_k were functions of a single spatial coordinate x . These quantities obey the continuity equation

$$\partial_t n_k(x, t) = \partial_x j_k + g_k(x, t) - n_k/\tau_k, \quad (7)$$

where τ_k is the lifetime of the excitons in k th CQW, which is known from the experiment, and g_k is the generation rate proportional to the local laser power. To represent the exciton currents, we used the drift-diffusion approximation,

$$j_k(x, t) = -D_k \partial_x n_k(x, t) - B_k n_k(x, t) \partial_x \mu_k, \quad (8)$$

where D_k and $B_k = D_k/T$ are the diffusion coefficient and the drift mobility, respectively. Finally, to simplify the treatment of interaction effects, we linearized the density dependence of the exciton chemical potentials, such that

$$\mu_1 = \gamma_{11} n_1 + \gamma_{12} n_2, \quad \mu_2 = \gamma_{22} n_2 + \gamma_{12} n_1, \quad (9)$$

where γ_{ij} are interacting constants. Based on the simulations presented in Figs. 4a and 4d of the main text, we set the constants to be $\gamma_{11} = 9.3$, $\gamma_{22} = 11$, and $\gamma_{12} = -0.2$, all in units of $10^{-11} \text{ meV} \cdot \text{cm}^{-2}$. [Note that the intra-CQW coupling constants are 1/3 of the ‘‘plate capacitor’’ values, i.e., $\gamma_{kk} = (1/3) \times (4\pi e^2 d_k/\epsilon)$]. We developed a computer program that solves these coupled equations on a discrete grid of x as a function of the time variable t , starting from initial conditions $n_1 = n_2 \equiv 0$. To get a relation between the laser powers and the generation rates, we fitted the shifts $E_k \approx \mu_k$ of the exciton emission energies measured as functions of the laser power to the results of these simulations. We estimated the diffusion coefficients D_k by fitting the calculated width of the IX_1 and IX_2 exciton clouds to the measured widths of these clouds generated selectively by L_1 or L_2 .

As outlined in the main text, the simulations show that the IX_1 cloud attracts to the IX_2 cloud (Fig. 5), in qualitative agreement with the attraction observed in the experiment (Fig. 3). In comparison, both our experimental measurements (Fig. 8) and theoretical simulations (Fig. 9) show that two clouds of repulsively interacting IX_2 repel each other.

APPENDIX C: POSITION-ENERGY LUMINESCENCE IMAGES

The differential x -energy image (Fig. 1b) is obtained by subtracting the x -energy images created by only L_1

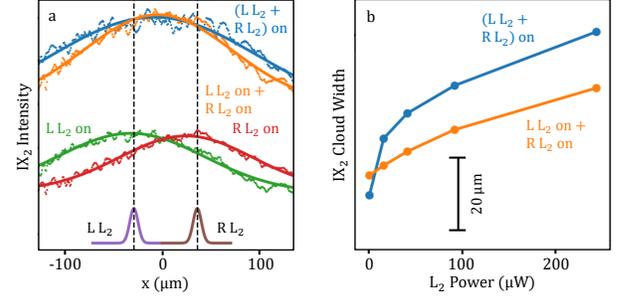


FIG. 8: Repulsion between the IX_2 clouds: Experiment. (a) The IX_2 cloud profiles when only left L_2 is on ($L L_2$ on, green line), when only right L_2 is on ($R L_2$ on, red line), and when both left and right L_2 are on [$(L L_2 + R L_2)$ on, blue line]. The sum of $L L_2$ on profile and $R L_2$ on profile ($L L_2$ on + $R L_2$ on) is shown by orange line. Profile $(L L_2 + R L_2)$ on is wider than the sum of $L L_2$ on profile and $R L_2$ on profile, indicating the repulsion between the IX_2 clouds. The profiles of left and right L_2 laser excitation spots are shown by purple and brown lines, respectively. Dashed lines indicate the centers of the excitation spots. (b) The width of IX_2 cloud when both left and right L_2 are on [$(L L_2 + R L_2)$ on, blue points] in comparison to the width of IX_2 cloud obtained as the sum of $L L_2$ on cloud and $R L_2$ on cloud [$L L_2$ on + $R L_2$ on, orange points] as a function of P_{L_2} .

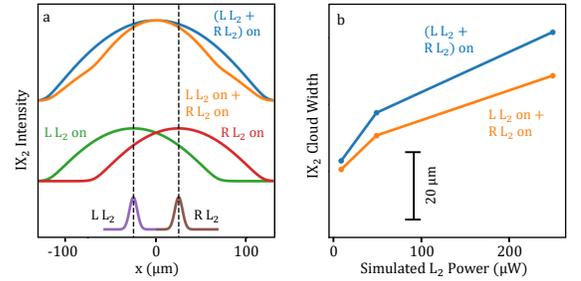


FIG. 9: Repulsion between the IX_2 clouds: Theory. (a) The IX_2 cloud profiles simulated when only left L_2 is on ($L L_2$ on, green line), when only right L_2 is on ($R L_2$ on, red line), and when both left and right L_2 are on [$(L L_2 + R L_2)$ on, blue line]. The sum of simulated $L L_2$ on profile and $R L_2$ on profile ($L L_2$ on + $R L_2$ on) is shown by orange line. Profile $(L L_2 + R L_2)$ on is wider than the sum of $L L_2$ on profile and $R L_2$ on profile, indicating the repulsion between the IX_2 clouds. The profiles of left and right L_2 laser excitation spots are shown by purple and brown lines, respectively. Dashed lines indicate the centers of the excitation spots. (b) The width of simulated IX_2 cloud when both left and right L_2 are on [$(L L_2 + R L_2)$ on, blue points] in comparison to the width of IX_2 cloud obtained as the sum of simulated $L L_2$ on cloud and $R L_2$ on cloud [$L L_2$ on + $R L_2$ on, orange points] as a function of P_{L_2} .

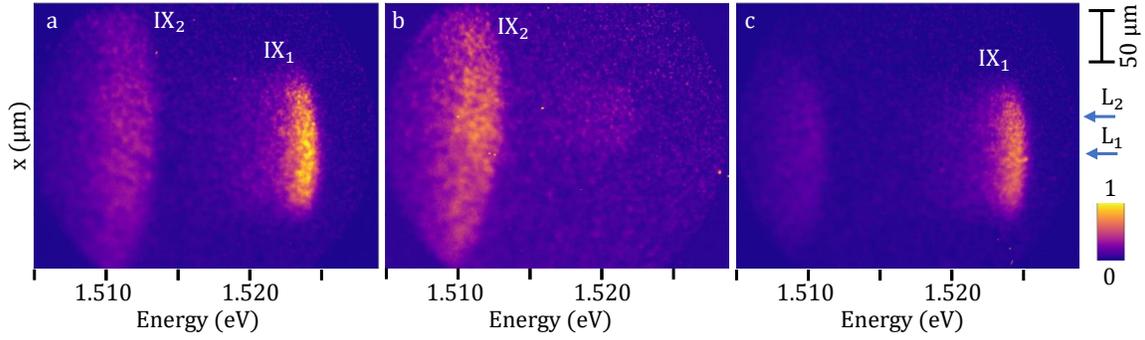


FIG. 10: **Position-energy images of IX luminescence.** (a) Both L_2 and L_1 lasers are on. (b) Only L_2 is on. (c) Only L_1 is on. The arrows indicate the excitation spot positions of L_2 and L_1 lasers resonant to direct excitons in 15 nm CQW and 12 nm CQW, respectively. L_2 generates IX_2 . L_1 generates IX_1 and also a smaller concentration of IX_2 . The laser powers $P_{L1} = 10 \mu\text{W}$, $P_{L2} = 250 \mu\text{W}$. Gate voltage $V_g = -2.0 \text{ V}$, temperature $T = 1.7 \text{ K}$.

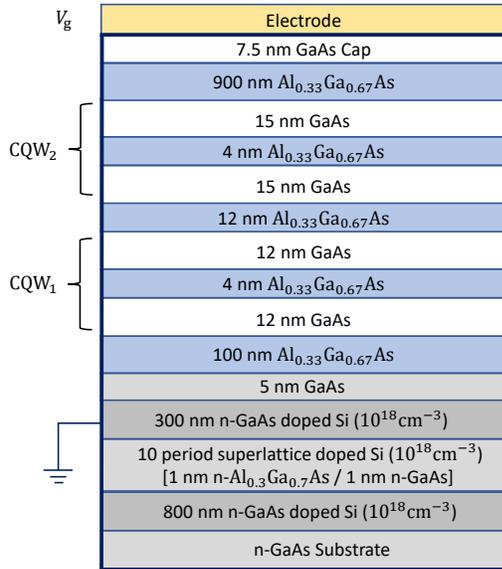


FIG. 11: **Cross-section of the 2-CQW heterostructure.** The thicknesses and doping concentrations of the layers are indicated.

on (Fig. 10c) and by only L_2 on (Fig. 10b) from the x-energy image created by both lasers on simultaneously (Fig. 10a).

APPENDIX D: CROSS-SECTION OF THE HETEROSTRUCTURE

The cross-section of the 2-CQW heterostructure showing the thicknesses and doping concentrations of the layers is presented in Fig. 11.

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