Detecting coupled excitons with microphotoluminescence techniques in bilayer quantum dots

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The coupling effects between excitons are examined in a pair of quantum dots to probe coupled excitons in optical measurements. The exciton photoluminescence (PL) and absorption spectrum are measured with the micro-PL excitation (micro-PLE) technique in bilayer InGaAs quantum dots. The PL peaks from the coupled exciton have similar PLE spectra for a weak excitation, which agree well with a theoretical prediction. Additional PL peaks appear for a strong excitation. A sum rule in four PL peaks is satisfied due to the energy conservation rule in the case of two exciton creation. A simple control of the coupling effect is also demonstrated using a two-color excitation technique. The coupling energy can be controlled by the intensity of a second laser. These results provide good guidelines for finding PL peaks from coupled excitons, which are crucially important to demonstrate scalable quantum gates and quantum computing with quantum dots.

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I. INTRODUCTION

Semiconductor quantum dots have been used to conduct many research issues because of their intriguing physics and possible novel device applications.[1,2] Recently, quantum dots have been attracting a lot of attention as a good candidate for future devices in quantum information processes. One such application is quantum communication. Nonclassical single photon emissions and quantum key distributions were demonstrated utilizing photoluminescence processes from excitons in isolated quantum dots.[3,4] Another important application is quantum computing.[5] Excitons created by optical pulses become quantum bits (qubits), which are basic units of quantum gates, and sequences of optical pulses control gate operations.

Quantum computing requires two kinds of fundamental gates. One is rotational gates with single qubits. The other is conditional NOT (CNOT) gates with two qubits. Fundamental aspects of the rotational gates were clarified in several quantum dot systems and controlling gates were achieved with optical pulse excitation techniques.[6–8] These works might lead to implementations of the rotational gates with quantum dots. In contrast, a demonstration of CNOT-like gate has been reported only by Li *et al.* using exciton-biexciton systems in single isolated quantum dots.[9] The exciton-biexciton systems provide good quantum correlation processes between an exciton and a biexciton which serve as two qubits. The quantum correlation processes between two qubits are necessary for CNOT gates. However, an exciton-biexciton system creates a quantum correlation process only between two predefined qubits. Even when many CNOT gates with exciton-biexciton systems exist, each gate acts as an independent gate; there may be no correlations among qubits. Usually, quantum computing processes calculation tasks utilize the quantum algorism. Most of the quantum algorism needs quantum correlations among many qubits. Such algorism requires CNOT gates where two qubits are appropriately selected from correlating many qubits. Although the CNOT gate with the exciton-biexciton system allows us to examine its fundamental properties, this gate is not suitable to demonstrate and implement quantum computing. These CNOT gates are not scalable to design actual quantum computers.

As for scalable quantum gates, coupled quantum dot systems have been proposed.[10,11] Coupled quantum dots give quantum correlated many exciton systems. The sources of the quantum correlations are the tunneling effect and the dipole-dipole interaction. The fundamental properties of the correlation effects have been clarified by theoretical considerations both for the tunneling and the dipole-dipole interaction.[12–15] Some studies have also proposed possible schemes of CNOT gates. In contrast, few experimental studies have been reported on the subjects of the coupled quantum dots[16–19] and CNOT gates have not yet demonstrated. Experimental verifications of the coupled quantum dots are not simple because there are no established guidelines to confirm coupled quantum dots while excluding the effects of uncoupled quantum dots. Usual experimental results such as photoluminescence (PL) spectra include PL peaks from coupled quantum dots as well as uncoupled quantum dots. Previous theoretical analyses focus on the properties of coupled quantum dots and do not provide sufficient information on such verifications. Clear guidelines for verifying coupled quantum dots experimentally will boost experimental studies as well as relevant theoretical works. Moreover, these are very useful to implement CNOT gates and demonstrate quantum computing.

In this paper, we examine the optical properties of coupled excitons between a pair of isolated quantum dots in a bilayer quantum dot sample to obtain guidelines for detecting coupled excitons. Previous reports assigned coupled excitons from PL energy variations in many quantum dots samples of different separations,[16] time-dependent properties,[17] and photon correlations.[19] Our guideline relies only on PL excitation (PLE) spectra and a sum rule in PL peak energies. We measured the PL and PLE properties of excitons in a bilayer InGaAs quantum dot sample by using the micro-PL technique. A simple theoretical consideration showed that PLE spectra for a pair of coupled excitons should have similar structure. We assigned two PL peaks for a pair of coupled excitons selected from other PLs for uncoupled excitons. When we increased excitation intensity, we found that other additional PL peaks appeared and several PL emission peaks strictly satisfy a kind of energy conservation rule. This result also agrees with the theoretical consideration. These results provide a good guideline for detecting coupled excitons. We also demonstrate a control of coupled exciton states by using the micro-PL with the two-color excitation method. The coupling energy can be tuned by changing laser intensity, which is a fundamental requirement of actual CNOT gates. Our results will be an important step for demonstrations of CNOT gates with coupled quantum dots.

II. EXPERIMENT

The InGaAs quantum dot sample for our measurement was fabricated on a GaAs (311) B substrate by metalorganic vapor phase epitaxy (MOVPE). The sample has two quantum dot layers and separation between each layer is about 5 nm. The fabrication process has already been reported in detail.[20,21] Measurements were performed using the micro-PL method.[22–24] The excitation laser light from a cw wavelength tunable Ti-sapphire laser was focused on the quantum dot sample with a microscope objective lens (spot size 2 μm) through a metal mask with holes processed on the sample surface (hole size 0.5 μm). For the two-color excitation, a cw diode pumped solid state laser (green laser) was used, whose wavelength was 532 nm. The sample was mounted on the cold head in a liquid He cooled cryostat. The PL was collected by the lens and detected by a liquid-nitrogen-cooled charge-coupled device through a high-resolution spectrometer. This method enables us to measure the PL properties from a few isolated quantum dots. PL measurement with the scanning wavelength of the excitation laser gives PLE spectra that directly reflect the photoabsorption spectra and energy structure of the quantum dot. We studied optical properties of coupled excitons from the PL and PLE spectra in a bilayer InGaAs quantum dot sample.

III. RESULTS AND DISCUSSION

Figure 1 shows an example of PL spectrum in the bilayer quantum dots taken at 4 K. The excitation density is about 800 W/cm^2 and the excitation energy is about 1.771 eV. Several PL peaks can be seen in the spectrum. Some PL peaks might be from coupled quantum dots and some PL might be from uncoupled ones. It is not so easy to distinguish the PLs of coupled quantum dots from those of uncoupled ones. This is because there are few guidelines to detect such PLs at the present. We consider the guidelines by using a simple theoretical analysis.

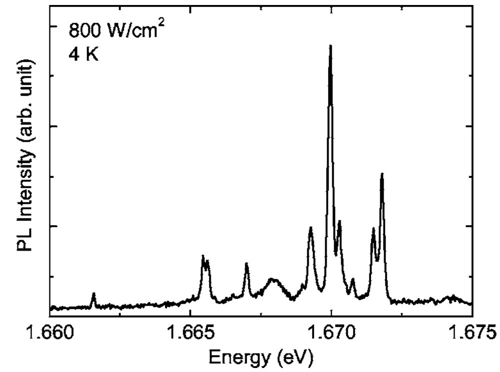

FIG. 1. PL spectrum for a bilayer quantum dot sample. The separation between a dot pair is about 5 nm. The excitation intensity is about 800 W/cm^2.

The Hamiltonian for two coupled quantum dots may be written as follows,[14]

$$H = \begin{bmatrix} g & 0 & 0 & 0 \\ 0 & E_1 & V_F & 0 \\ 0 & V_F & E_2 & 0 \\ 0 & 0 & 0 & E_1 + E_2 - E_b \end{bmatrix}, \quad (1)$$

where E_i $(i=1,2)$ are exciton ground energies for two quantum dots, g is the ground state energy, V_F is the coupling energy between quantum dots and E_b is the interaction energy for two dot excitation. Here, V_F is for the tunneling energy or the dipole-dipole coupling energy. When excitation intensity is in the weak region, only a single exciton is created and the system may be a three-level system. In Eq. (1), only upper 3×3 elements should be considered. The coupling effect between two exciton states results in two new states that are expressed by a linear combination of original wavefunctions.

Figure 2(a) shows an energy level diagram of two (A and B) quantum dots for a weak excitation. $A0$ and $B0$ are the lowest exciton states for two quantum dots. The state g is the ground state. The wavefunctions may be represented by $|10\rangle$, $|01\rangle$, and $|00\rangle$, respectively. When the $A0$ and $B0$ states are coupled, these states form new states (α and β). When the energy of $A0$ is equal to that of $B0$, the wavefunctions of α and β are $\frac{1}{\sqrt{2}}(|10\rangle - |01\rangle)$ and $\frac{1}{\sqrt{2}}(|10\rangle + |01\rangle)$, respectively. These new states have the characteristics of the original two dots. In Fig. 2(a), $A1$, $A2$, $B1$, and $B2$ are higher exciton states for two quantum dots. Here, we assume that the coupling effects exist only between $A0$ and $B0$; no coupling occurs in these higher states. The higher exciton states can be probed by PLE measurements which give us absorption spectra of quantum dots.[22–24] When an exciton is created through states of $A1$ or $A2$, the exciton relaxes into the ground state $A0$. The relaxed exciton couples with $B0$ state

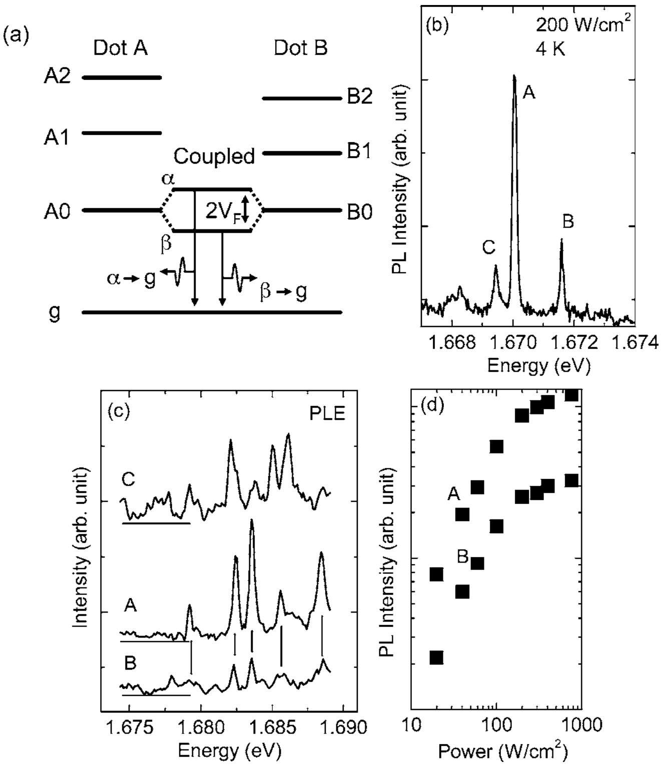

FIG. 2. Energy-level diagram for coupled excitons for a weak excitation. Only a single exciton is created in dot A or B. Coupling occurs only between $A0$ and $B0$ states which are the lowest exciton states. (b) PL spectrum for a weak excitation (200 W/cm^2). (c) PLE spectra for PL peaks A, B and C in (b). (d) Power dependence of PL peaks A and B.

forming new α and β states. In contrast, a relaxed exciton in $B0$ state from $B1$ or $B2$ couples with $A0$ resulting in the formation of α and β states. Therefore, all created exctions from higher states in both dots A and B can emit PLs from both the α and β states. This means that two PL emission peaks for $\alpha \rightarrow g$ and $\beta \rightarrow g$ should have similar PLE spectra. Energy difference between two PLs is twice the coupling energy V_F. These characteristics are keys for selecting PL peaks from coupling quantum dots.

Figures 2(b) and 2(c), respectively, show PL and PLE spectra in a bilayer InGaAs quantum dot sample. Three PL peaks can be seen in Fig. 2(b). The structure of the PLE spectrum for peak A is similar to that for peak B in Fig. 2(c). However, the PLE spectrum for peak C has a different structure from the other two spectra. These results imply that PL peaks A and B are generated from coupled excitons. These PLE spectra indicate that PL peaks A and B originate from a pair of coupled exciton in coupled quantum dot and that PL peak C is from an uncoupled, isolated exciton. However, there is another possibility for the similar PLE structures. We reported that an exciton and a biexciton in a quantum dot have similar PLE spectra.[23,24] This characteristic does not require any coupling effects. We consider this possibility from excitation intensity dependence of PL peaks A and B. Figure 2(d) shows PL intensity for PL peaks A and B as a function of excitation intensity. The PL intensities for both peaks increase linearly up to 200 W/cm^2 and saturate above this value. When the PL is from an biexciton, the PL intensity should increase with the square of the excitation intensity. The exciation dependence in Fig. 2(d) rules out the biexciton contribution to the PL peaks. The result of Figs. 2(c) and 2(d) strongly suggest that PL peaks A and B in Fig. 2(b) is from a pair of coupled quantum dots.

We explore another guideline for finding PLs from coupled quantum dots. Figure 3(a) shows an energy-level diagram for a strong excitation. In Eq. (1), all 4×4 elements should be considered for this condition. The diagram has a two-exciton state ($AB0$) where excitons are created in both dot A and dot B. E_b is the binding energy for closely existing two excitons, whose origin is similar to the biexciton binding energy. With a strong excitation, four PL emission processes can be considered. Four PL energies are different from each other due to the finite binding energy E_b. When the excitons are created in $AB0$, this state has two paths to reach the g state, and each path includes two PL emission processes. Figure 3(a) clearly shows that the energy conservation rule is satisfied in consecutive PL processes from $AB0$ to g; $E_{AB0\rightarrow\alpha}+E_{\alpha\rightarrow g}=E_{AB0\rightarrow\beta}+E_{\beta\rightarrow g}$. This characteristic constitutes another proof of the existence of coupled excitons. Figure 3(b) plots the PL spectrum in the same dot for a strong excitation condition. Some additional PL peaks appear that were not seen in Fig. 2(b). We focus on the peaks denoted D and E. The sum of the PL energies for peak A and D is 3.34176 eV. This value is equal to the total for peaks E and B. This clearly shows that peaks D and E are both PL emissions from the $AB0$ state, and correspond to $AB0 \rightarrow \beta$ and $AB \rightarrow \alpha$, respectively. These PL characteristics support that

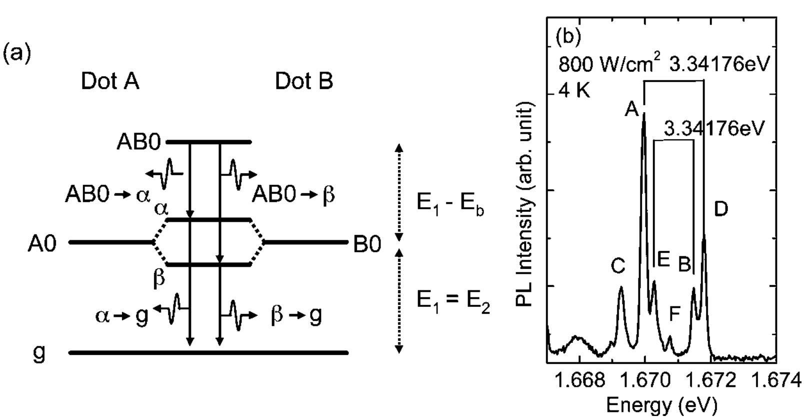

FIG. 3. Energy-level diagram for coupled excitons under a strong excitation. This include the state $AB0$ where excitons exist in both dots. (b) PL spectrum for 800 W/cm^2. The energy range is the same as that in Fig. 2(b). Additional PL peaks can be seen.

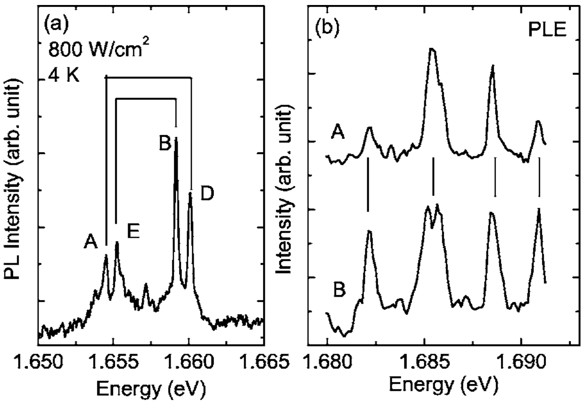

FIG. 4. PL spectrum for a strong excitation (800 W/cm^2) for a different quantum dot from Figs. 2 and 3. (b) PLE spectra for PL peaks A, B in (a).

PL peaks A and B are from a pair of coupled excitons and become another key to detecting such excitons. Our results for PLE and PL characteristics provide clear and simple guidelines for detecting PL emissions originated from coupled excitons while excluding the effects of other PL lines for uncoupled excitons. These guidelines rely only on PL and PLE spectra in a bilayer quantum dot sample. These are useful to detect the effects of coupled excitons.

Here we consider the density of the coupled quantum dots. The quantum dot density is about 10^9–10^{10} cm^{-2}.[20,21] There are about 3–30 quantum dots per 0.5 μm^2 hole. Currently, the percentage of coupled quantum dots is estimated to be typically 10–20%. We showed the PL and PLE spectra for other quantum dots in Fig. 4. The PL and PLE energy regions are different from those in Figs. 1 and 2. This may be due to dot size and indium content variations in the quantum dot sample. We found several coupled quantum dots with similar PL and PLE spectra from every 20–30 quantum dots. In Fig. 4, the sum of the PL energies for peaks A and D is about 0.2 meV larger than that for peaks E and D. The energy difference is close to the linewidth of each PL. This result may be due to additional coupling effects induced by other quantum dots. In quantum dots, coupling effects may exist in the vertical direction as well as the lateral direction. These coupling effects with other quantum dots, which are much smaller than the main coupling effects, can change the PL energy as small as PL linewidths. This small deviation in the PL energy sum rule can be seen in some quantum dots.

We discuss the physical origin of the coupling. Two mechanisms are proposed for the coupling; the tunneling effects and the dipole-dipole interactions.[16–19] The experimental method of distinguishing these two effects has not yet been established. Observation of anticrossing effects[16] and the photon correlation measurement[19] might be possible ways. Here we compare the coupling energy estimated from PL spectra with theoretical predictions. Recent theoretical calculation concludes that the tunneling effects are suppressed between nonidentical quantum dots even for the several nanometer separation.[15] Therefore, we consider the dipole-dipole interactions. Nazir *et al.* reported Föster interaction strength as a function of dot separations for various dipole moments.[14] Föster interaction is the one dipole-dipole interaction that has dominant effects for the dot separation of less than about twenty nanometers. The Föster energies for 5 nm separation are about 1 meV for the dipole moment of 33 Debye and 0.5 meV for 24 Debye. The coupling energy estimated from Fig. 2(b) is 0.8 meV (half of the energy between peak A and B). The dipole moment of our dot is about 30–50 Debye.[7] The coupling energy is very close to the results of the calculation. This suggests that the coupling effects in our quantum dots are due to the dipole-dipole coupling. Confirmation of the mechanism may require other measurements, and this issue may be considered in the future.

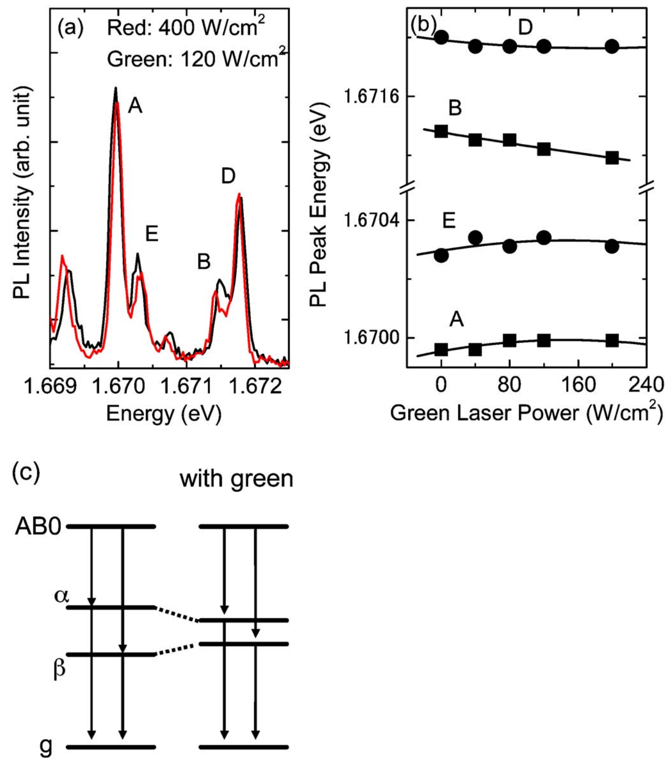

FIG. 5. (Color online) (a) PL spectra for the single- and two-color excitation conditions for the quantum dots in Figs. 2 and 3. PL peaks shift with the green laser excitation as shown by the red (dark gray) line. (b) PL energies as a function of the intensity for the green laser. (c) Energy-level diagram for single- and two-color excitations.

The coupling effects between two quantum dots strongly depend on several physical parameters such as the energy difference between two exciton states, the coupling energy and separation between two quantum dots according to reported theoretical and experimental studies.[13–19] PL peak energies from coupled excitons will change with these parameters. Tuning these parameters by some methods are crucially important to achieve schemes for quantum gating with coupled excitons. We examined PL characteristics for the two-color excitation. The additional green laser creates excitons in barrier regions around the coupled quantum dots. These excitons may influence coupling effects between the quantum dots and thereby change the coupling energy. These effects can be evaluated from PL spectra. Figure 5(a) shows PL spectra for the single (red) and two-color (red and green) excitation conditions for the quantum dots in Figs. 2 and 3. Excitation conditions for the single laser are the same as those in Fig. 3. By illuminating the green laser, many PL peaks are shifted in energy. While we increased only the intensity of the red laser, no PL shifts were observed (not shown). We focus on the four PL peaks that originate from coupled quantum dots as discussed above (peaks A, B, D, and E). Figure 5(b) displays PL peak energies as a function of the intensity of the green laser. With increasing green laser intensity, PL peaks A and E blueshift and peaks B and D redshift. These results are due to changes in the coupling energy induced by excitons in the barrier regions. An energy-level diagram is shown in Fig. 5(c). This diagram explains changes in PL energy when the coupling energy is decreased. Decreasing the coupling energy causes blueshifts in two PL peaks and redshifts in the other two PL peaks. The results in Figs. 5(a) and 5(b) can be interpreted in terms of decreasing in coupling energy. The excitons in barrier regions may induce additional dipole-dipole coupling effects and Coulomb effects, which lead to decreasing in the coupling energy.

In Fig. 5(b), the sum of the PL energies B and E is equal to that for A and D without the green laser. This relation is not satisfied for the strong excitation region. Although the reason for this is yet unclear, one possibility is the effect of the additional dipole coupling induced by green laser excitation. This implies intriguing physics where the additional dipole coupling is dominant only for one quantum dot but not for both coupled quantum dots. The physics of dipole coupling between quantum dots and barriers is not yet fully understood and will be reconsidered in the future.

The characteristics in the measurement for Fig. 5 ensure again that four PL peaks are from coupled quantum dots. Moreover, this experiment provides a possible scheme of controlling the properties of coupled quantum dots and may constitute an important milestone for demonstrating CNOT gate operations.

The CNOT gate, which is a fundamental requirement for the quantum computing, may be demonstrated in coupled quantum dots. This demonstration requires detailed evaluations of the quantum correlation between two qubits and sophisticated controlling schemes. Implementing quantum computing needs correlated many qubits and quantum gates. Previously reported CNOT in quantum dots is substantially a single isolated CNOT gate and not easy to apply to the quantum computers. A coupled quantum dot can also couple to other coupled quantum dots through dipole-dipole coupling. This scalability is very useful for achieving demonstrations of quantum computing in quantum dots.

IV. CONCLUSION

We have investigated coupling effects between two excitons on PL characteristics in bilayer quantum dots. PL and PLE spectra were measured in a bilayer InGaAs quantum dot sample by using a single-dot spectroscopy. Two guidelines for finding PL peaks from a pair of coupled excitons were shown using PLE spectra and the sum rule in PL peak energies. Several PL peaks are seen in a PL spectrum in the quantum dot for a weak excitation condition. Two PL peaks that had similar PLE structures were from a pair of coupled excitons which agrees well with a simple theoretical consideration. For a strong excitation, additional PL peaks appeared, and a sum rule in energies of four PL peaks is satisfied. These guidelines are very useful for finding PL peaks for coupled excitons separating those for uncoupled excitons. We also showed a simple demonstration of controlling coupling effects using a two-color excitation technique. The coupling effects are tuned by changing the intensity of the green laser. This demonstration provides a good milestone to implement scalable CNOT gates. Our result will assist us in making progress in detecting and controlling coupled excitons and will be useful when applied to quantum computing with quantum dot excitons.

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