

# Exciton fine structures and energy transfer in single InGaAs quantum-dot molecules

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We present a spectroscopic study of single quantum-dot molecules (QDMs) formed by two closely stacked In<sub>0.5</sub>Ga<sub>0.5</sub>As/GaAs layers. It was found that the interdot coupling and directional energy transfer between the two adjacent dots can be controlled by temperature tuning. Direct and indirect excitons, as well as charged excitons in single QDMs were classified and identified by excitation-power dependent, excitation-energy dependent and polarization-resolved micro-

photoluminescence measurements. With the increasing temperature, the direct-exciton intensity decreases while the indirect-exciton intensity increases. A rate equation model considering phonon mediated processes has been developed. The directional energy transfer in QDMs is explained in terms of the phonon-assisted tunnelling of hole between the two adjacent dots.

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**1 Introduction** The underlying atom-like properties of single isolated semiconductor quantum dots (QDs) have proven to be very useful for the implementation of various quantum information applications, including quantum cryptography using single QDs as non-classical light sources [1–4] and quantum gate [5] operation based on exciton excitations. Building molecular-like structures with controllable coupling effect provides a possible route for further scalability of such applications. Recent experiments have revealed that electrical control of tunnel coupling is feasible in single QD molecules (QDMs) formed by a pair of either vertically stacked [6–9] or laterally aligned [10] InGaAs self-assembled QDs. Despite the success in explaining the rich pattern of level anticrossings/crossings in accordance with an applied electric field, the spectral features of a single QDMs is by far more complicated than their single QD counterparts, since QDMs are usually made of two non-identical dots and the coupling mechanism will also differ with the interdot separation. The directional energy transfer of exciton between the two adjacent dots was observed and

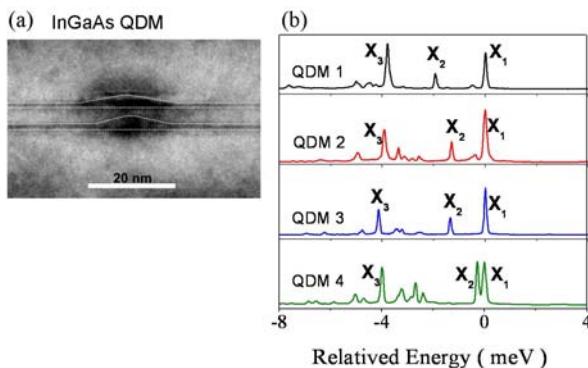
explained in terms of phonon-assisted Förster processes [11, 12]. However, such a directional transfer of carriers could also arise from nonresonant tunneling, which has been reported on ensemble of In(Al,Ga)As asymmetric QD pairs [13, 14], and recently on single InP/InGaP QD pairs [15]. Therefore, it is important to clarify whether the Förster transfer or the nonresonant tunneling is responsible for the directional energy transfer in QDMs.

In this work, we present a spectroscopic study of single QDMs formed by two closely stacked In<sub>0.5</sub>Ga<sub>0.5</sub>As/GaAs QD layers. Fine structures of direct and indirect excitons in QDMs were investigated by power dependent and polarization resolved micro-photoluminescence ( $\mu$ -PL) measurements. A directional energy transfer from a direct to an indirect exciton in single QDMs was found as the temperature was increased. This phenomenon is explained in terms of a thermally activated tunneling of the hole between the two adjacent QDs. In this work, we present a spectroscopic study of single QDMs formed by two closely stacked In<sub>0.5</sub>Ga<sub>0.5</sub>As/GaAs QD layers [16].

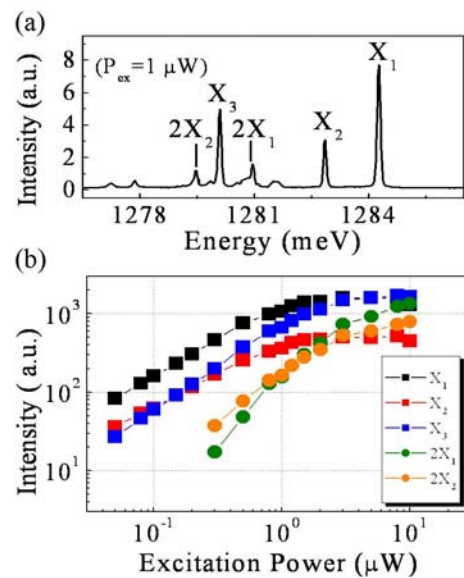
**2 Experimental** Our QDMs samples were grown by metalorganic chemical vapour deposition (MOCVD), yielding a low QD density of about  $108\text{--}109\text{ cm}^{-2}$  by a careful control of the InGaAs coverage [17]. The layer sequence consists of a 100-nm undoped GaAs buffer layer, followed by a 500-nm  $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$  layer and a 80-nm GaAs layer grown at  $700\text{ }^\circ\text{C}$ . The QDMs, formed by a pair vertically stacked  $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$  QD layers, separated by a thin GaAs spacer layer, were then grown at  $500\text{ }^\circ\text{C}$ . Cross-sectional transmission electron microscopy (TEM) reveals that the InGaAs QDs in each layer is lens-shaped, and about 3 nm in height situated on a 1.8-nm-thick wetting layer (WL) as shown in Fig. 1(a). The two WLs are separated by a 5-nm-thick GaAs spacer layer, corresponding a tip-to-base distance of only  $\sim 2$  nm. Individual QDM spectra were investigated by a  $\mu\text{-PL}$  setup via an Al metal mask with arrays of nano-apertures. A He-Ne laser beam was focused on the aperture via a microscope objective (N.A. = 0.5,  $100\times$ ). The PL signals were analyzed by a 0.75 m grating monochromator combined with a liquid-nitrogen-cooled CCD camera, which yields a resolution limited spectral linewidth of  $\sim 60\text{ }\mu\text{eV}$ . By using the Lorentzian line-shape fitting, the peak position of emission lines can be determined with an accuracy better than  $10\text{ }\mu\text{eV}$ .

**3 Results and discussion** We have investigated a number of single QDMs to date, and most of which show similar behaviors. Typical  $\mu\text{-PL}$  spectra taken from four different QDMs excited at 1.96 eV under  $1\text{ }\mu\text{W}$  are displayed in Fig. 1(b). Under low excitation conditions, three dominant emission lines (labelled  $X_1$ ,  $X_2$ , and  $X_3$ ) were invariably observed. The  $X_1$  and  $X_2$  lines are separated by an energy varying from 0.3 to 2 meV for different QDMs, while the  $X_3$  is invariably present at an energy of about  $4.0 \pm 0.3\text{ meV}$  below the  $X_1$  line.

Power-dependent PL measurements have been performed in order to classify these emission lines. A typical spectrum obtained from QDM3 is shown in Fig. 2(a). The dependence of PL intensity ( $I_{\text{PL}}$ ) on the excitation power



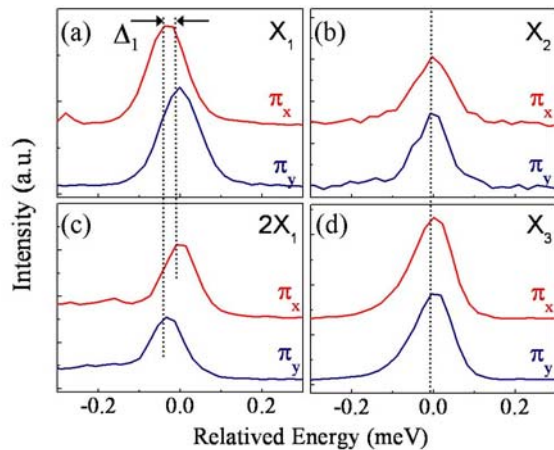
**Figure 1** (a) Cross sectional TEM image of the InGaAs QDM structure. (b) PL spectra taken from different QDMs. The energy scale is relative to the  $X_1$  peak energy at 1257.4, 1285.3, 1284.3, and 1283.8 meV from QDM1 to QDM4, respectively.



**Figure 2** Power-dependent  $\mu\text{-PL}$  spectra of QDM3 at excitation energy = 1.96 eV. (a) The PL spectrum taken at excitation power =  $1\text{ }\mu\text{W}$ . (b) The integrated PL intensity of each emission line as a function of excitation power.

( $P_{\text{ex}}$ ) for each line can be characterized by  $I_{\text{PL}} \propto P_{\text{ex}}^\alpha$ . As shown in Fig. 2(b), both the  $X_1$  and  $X_2$  intensities increase linearly with  $P_{\text{ex}}$ , with exponents of 1.0 and 0.95, respectively. We therefore identify  $X_1$  and  $X_2$  as different neutral excitons state in the QDM. The  $X_3$  line shows a superlinear dependence on  $P_{\text{ex}}$ , with an exponent of  $\sim 1.3$ , which can be ascribed to the negatively charged exciton ( $X^-$ ). The formation of  $X^-$  state is related to the unintentionally doped carbon impurities in the MOCVD grown sample, leading to preferential captures of more electrons into the QDM under nonresonant excitations [18]. Here, it is important to point out that the energy separations between  $X_1$  and  $X_3$  are almost the same for different QDMs. Because the binding energy of  $X^-$  is less sensitive to the dot size, the similar binding energy implies that the  $X_3$  line is the negatively charged state of the  $X_1$  line in the QDM. Another two peaks marked as  $2X_1$  and  $2X_2$  were also analyzed. Their quadratic and superlinear power dependencies indicate that they are recombinations from of biexciton states.

The  $X_1$  and  $X_2$  emission lines are unlikely to arise from direct excitons localized in the two different dots of the QDMs, because the inherent size difference of two dots would cause a difference in ground state energy of tens of meV, which cannot account for the variation in  $X_1$ - $X_2$  separation of only a few meV's. For QDMs with spacer thickness as thin as 5 nm, theoretical calculations predicted that the lowest lying two electron states hybridize into bonding and anti-bonding orbitals with an energy splitting up to  $\sim 50\text{ meV}$  due to the strong interdot tunnel coupling. However, the lowest lying two hole states, which are split by only a few meV, remain essentially uncoupled even in such closely stacked QD pairs, due to the much larger hole



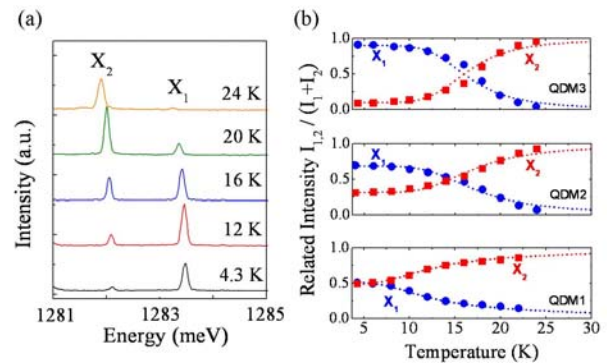
**Figure 3** Polarization-resolved PL spectra of QDM3.

effective mass and the interpenetrated strain field [19]. Therefore, we identify the two  $X_1$  and  $X_2$  lines as the recombination of the same electron state with two hole states localized in different dots, which can be characterized as a direct and an indirect transitions.

We have performed polarization-resolved PL measurements to further examine the direct and indirect natural properties of these emission lines based on the analysis of fine structure splitting (FSS) of exciton states in QDMs. In single dots, because the electron-hole ( $e-h$ ) exchange interaction is sensitive to the dot shape symmetry, the neutral exciton line will split into a linearly cross-polarized doublet [20]. For a QDM, if a direct exciton is localized in one particular dot of the QDM, the FSS is expected to resemble the single QD case. Furthermore, since the  $e-h$  exchange interaction is proportional to the overlap of the electron and hole wavefunctions, the FSS of the indirect exciton would be suppressed due to less wavefunction overlap.

In Fig. 3, linearly polarized spectra of QDM3 along  $[110]$  ( $\pi_x$ ) and  $[1\bar{1}0]$  ( $\pi_y$ ) directions are displayed. The  $X_1$  line consists of a linearly cross-polarized doublet with a FSS of  $\Delta_1 \sim 30$   $\mu\text{eV}$ . The FSS of  $2X_1$  is the same as that of  $X_1$ , but with a reversed polarization sequence, indicative of a cascade process for the direct exciton and biexciton in the same dot of the QDM. The  $X_3$  line does not show any FSS, as expected for an  $X^-$  state with singlet spin configuration of two electrons. In particular, we found that the FSS of the  $X_2$  line is virtually zero within our detection limit for all investigated QDMs. This leads us to believe that  $X_2$  is the indirect transition.

Temperature-dependent PL measurements have been performed in order to obtain more information about the interdot coupling. The temperature-dependent PL spectra of QDM3 were shown in Fig. 4(a). With the increasing temperature, we found that the  $X_1$  intensity ( $I_1$ ) decreases while the  $X_2$  intensity ( $I_2$ ) increases with a crossing in relative intensities  $I_{1,2} / (I_1 + I_2)$  at about  $T = 16$  K. All the investigated QDMs show a similar crossing behavior as shown in Fig. 4(b), but with different crossing temperatures. Be-



**Figure 4** (a) Temperature dependent PL spectra of QDM3. (b) The relative intensities of  $X_1$  and  $X_2$  transitions of different QDMs. The dashed lines are fitting curves calculated from the rate-equation model.

cause  $X_1$  and  $X_2$  are direct and indirect transitions, the intensity crossing indicates a directional transfer of hole between the two adjacent dots.

To understand the underlying transfer processes, a simplified rate-equation model considering an interdot transfer rate  $\gamma(T)$  from  $X_1$  to  $X_2$  was used. For simplicity, biexciton states are neglected in this model, which is applicable under low-excitation conditions. The thermally activated transfer rate was phenomenologically assumed to be  $\gamma(T) = \gamma_0 \exp(-E_A / k_B T)$ , where  $\gamma_0$  is a pre-exponential factor,  $E_A$  is the activation energy for the transfer process. By solving the rate equation in steady state, the relative intensity of  $X_1$  is given by

$$\frac{I_1}{I_1 + I_2} = \frac{g_1 / (g_1 + g_2)}{1 + (\gamma_0 / \gamma_1) \exp(-E_A / k_B T)}, \quad (1)$$

where  $g_1$  and  $g_2$  are generation rates of the  $X_1$  and  $X_2$  states, and  $\gamma_1$  is the recombination rate of the  $X_1$  state. The relative intensity of  $X_2$  can then be obtained from  $1 - [I_1 / (I_1 + I_2)]$ . Fitting parameters are  $g_1 / g_2$ ,  $\gamma_0 / \gamma_1$  and  $E_A$ , which determine the intensity ratio  $I_1 / I_2$  at low temperature, the crossing temperature, and the slope of intensity variation with temperature. As shown in Fig. 4(b), the simplified model well reproduces our experimental data. Inspections of several QDMs indicated that  $E_A$  varied in the range of 3–10 meV, without any correlation with their  $X_1$ - $X_2$  energy separation.

The observed transfer processes are unlikely to arise from resonant tunneling between the two adjacent dots, because the energy levels of nonidentical dots are usually not aligned. However, because excited states of the hole in QDs are less confined, tunnel coupling between hole excited states in the QDM will be more significant. Therefore, it is likely that the observed directional transfer of hole between the two dots is a thermally-activated tunneling, i.e., the hole in one dot first absorbed thermal energy (acoustic phonons) and activated to a higher-lying hole level, then tunneled into another dot, followed by rapid relaxations into the ground hole state to form an indirect exciton  $X_2$ .

For a square tunneling barrier, the interdot tunneling rate can be approximated by  $\gamma_{\text{tun}} \propto \exp[-2d\sqrt{2m_h\Delta V/\hbar^2}]$ , which increases exponentially with the decreasing barrier height  $\Delta V$  and thickness  $d$ . This explains why the interdot hole transfer can be considerably enhanced by absorption of thermal energy to available higher-lying levels.

**4 Conclusion** In summary, we presented a spectroscopic study of single QDMs formed by two closely stacked  $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$  QDs. The exciton fine structures as well as direct and indirect excitonic species associated with QDMs were identified by power dependent and polarization resolved micro-photoluminescence measurements. As temperature increasing, a directional energy transfer between the direct and indirect excitons in single QDMs was observed. A rate equation model was developed to explain our data. A thermally-activated tunneling of a hole between the two adjacent dots is responsible for such directional energy transfers in QDMs.

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