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Effects of p-type doping on the optical properties of InAs/GaAs quantum dots

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ABSTRACT

The effects of p-type doping on the optical properties of self-organized InAs/GaAs quantum dots (QDs) were investigated by both micro-photoluminescence and degenerated pump–probe reflection measurements. As compared to undoped InAs/GaAs QDs, it was observed that the transitions between the ground and the first excited states of electrons and holes levels appeared at higher energies for p-doped InAs/GaAs QDs. In addition, the PL intensities for both undoped and p-doped QDs were found to decrease when the excitation power exceeded a critical value. The critical excitation power for p-doped QDs appeared to be much lower than that for undoped ones. In the pump–probe experiments, it was revealed that the value and sign of the differential reflectivity depends strongly on excitation wavelength. P-doped QDs exhibited a response behavior that is different from that of undoped ones. It is believed that the large build-in population of holes plays a crucial role in determining the transient reflection spectrum.

1. Introduction

In the past two decades, semiconductor quantum dots (QDs) self-organized in the Stranski–Krastanov (SK) growth mode have been extensively studied due to their potential applications in various optoelectronic devices [1,2]. In particular, $In_xGa_{1-x}As$/GaAs QDs have received intensive studies because their operating wavelength can be designed at 1.3 μm which is suitable for optical telecommunication. Accordingly, much effort has been devoted to the investigation of the optical properties of self-organized QDs and especially carrier dynamics that may significantly affect the performance of QD-based devices. In general, the carrier dynamics in self-organized QDs include the capture of photogenerated carriers from the barrier and/or the wetting layer into QDs [3,4], the relaxation of carriers within QDs from the excited states to the ground state [5–7], and finally the recombination of carriers in QDs [8]. While in quantum wells fast carrier capture and relaxation are mediated by the carrier–phonon interaction, the carrier relaxation in QDs via phonon scattering is highly improbable due to their discrete energy levels. In particular, the capture and relaxation processes have always been the focus of the studies due to the worry of the phonon bottleneck effect predicted from the discrete energy levels of QDs [9,10]. From the viewpoint of device application, appropriate doping of QDs is necessary for making QD infrared photodetectors and a significant improvement in the modulation speed of QD lasers can be achieved by doping QDs [11–15]. Therefore, it is essential to clarify the effects of doping on the optical properties of self-organized QDs.

Micro-photoluminescence (μ-PL) is a popular technique that is used to characterize the optical properties of QDs. One advantage of this technique is high carrier densities achieved by focusing laser beam with an objective lens. As a result, the high-energy excited states of the QDs to be studied can be easily identified due to the state filling effect. The energy positions of these excited states can be extracted from the PL spectra under high excitation densities with appropriate Gaussian fitting. In addition, μ-PL can be employed to resolve the emission from single QDs when the area density of QDs is intentionally made to be low.

For the characterization of carrier dynamics in semiconductors, ultrafast optical techniques are widely applied and they rely either on the detection of transient photoluminescence (PL) [16,17], or on the measurement of differential transmission or reflection by pump–probe technique [3,18,19]. Although the time-resolved PL measurements provide the information of carrier capture and relaxation to the ground states of QDs, clear PL signals can only be obtained at low temperatures. In comparison, the pump–probe measurements can be carried out at room temperature and they offer good signal-to-noise ratio. This advantage makes them quite attractive for investigating the carrier dynamics in self-organized QDs. For a QD heterostructure, although the QD layer sandwiched by the buffer and cap layers is very thin, it plays an important role in determining the reflection of the QD heterostructure. In this article, we present a detailed comparison of the optical properties of undoped and p-doped InAs/GaAs QDs by utilizing both μ-PL and degenerate pump–probe reflection measurements.

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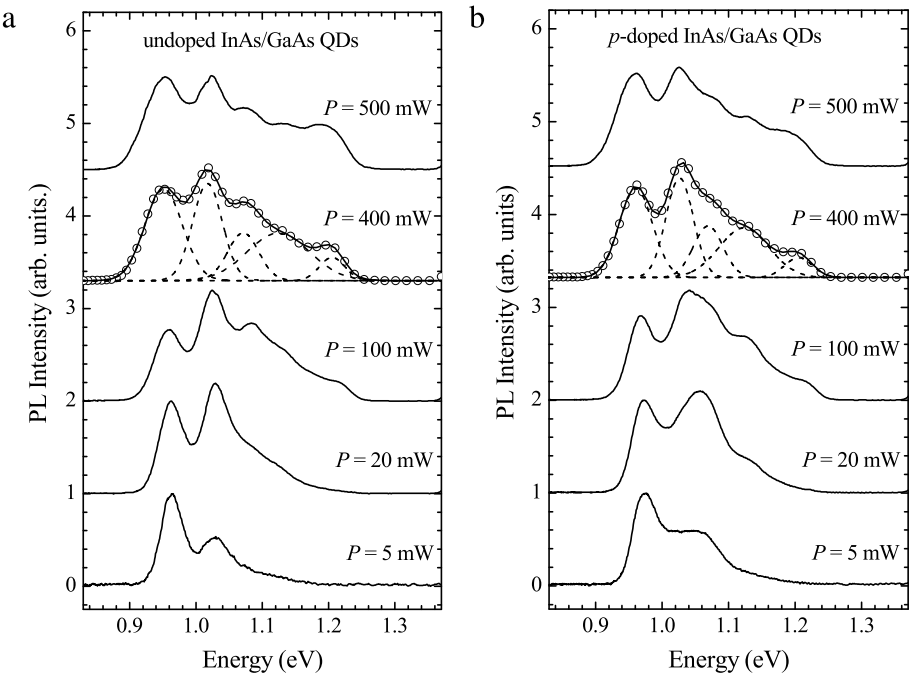

Fig. 1. PL spectra of (a) undoped InAs/GaAs QDs and (b) p-doped InAs/GaAs QDs under different excitation powers. Gaussian fittings for the PL spectra obtained at 400 mW are presented for both samples.

2. Experimental details

The undoped and p-doped InAs/GaAs QD samples used in the experiments were grown by molecular beam epitaxy. In both samples, InAs QDs were grown by the S–K mode on a 500 nm GaAs buffer at 500 °C. Then, they were covered by a 6 nm InGaAs strain relaxation layer followed by a 100 nm GaAs cap layer. On top of the GaAs cap layer, another layer of InAs QDs was deposited under the same growth condition for atomic force microscope (AFM) observation. According to the AFM images, the diameter and height of InAs QDs were measured to be ∼30 and 5–6 nm. In addition, the area density of InAs QDs was estimated to be 4×10^{10} cm^{-2}. For the p-doped QDs, a Be-doped GaAs layer (∼10 nm) was inserted into the GaAs cap layer. The doping density was chosen to be 10^{18} cm^{-3} which corresponds to ∼10 holes per QD. In order to find out the effects of *p*-doping on the optical properties of InAs/GaAs QDs, we have performed μ-PL and degenerate pump–probe reflection measurements for both QDs samples.

In μ-PL experiments, the 532 nm light from a solid-state laser (Verdi-5, Coherent) was introduced into an inverted PL microscope (Observer A1, Zeiss) and focused on the QD samples with a 100× objective lens. In this case, the diameter of the laser spot on the samples was estimated to be 1 μm. The PL was collected with the same objective lens and dispersed by a 0.3 m spectrometer with a spatial resolution of 0.1 nm. An InGaAs detector connected to the spectrometer was employed to record the PL spectra of the QD samples in a wavelength range of 1000–1500 nm. In order to resolve the discrete energy levels in the QD samples, we have gradually increased the pump power and recorded the PL spectra of the QD samples under different power densities.

The pump–probe reflection measurements were carried out at room temperature in a degenerate geometry by using a Ti:sapphire femtosecond (fs) oscillator (Mira 900, Coherent) pumped with a solid-state laser (Verdi-5, Coherent). The width and repetition rate of the fs laser pulse are 130 fs and 76 MHz. The wavelength of the fs laser pulses was tunable from 760 to 840 nm. At a wavelength of 800 nm, the energy of photons is about 130 meV above the absorption edge of GaAs. In the pump–probe experiments, the pump and probe beams were focused to spots of 150 and 50 μm in diameter and they are incident on the sample surface at angles of 0° and 45°, respectively. The time delay between them was generated by a delay line with a minimum step of 3 μm, which corresponds to a time resolution of ∼20 fs. The power of the probe beam was chosen to be 6 mW while that for the pump beam was set to be 120 mW.

3. Results and discussion

3.1. Effects of p-type doping on the energy states of QDs

The normalized PL spectra of the two QD samples with increasing excitation power (*P*) are shown in Fig. 1(a) and (b), respectively. At a low excitation power of 5 mW, only two peaks are observed in the spectra of the two QD samples. For excitation powers higher than 100 mW, one can clearly resolve five peaks in the spectra. The appearance of PL peaks at higher energies is attributed to the recombination of carriers at high-energy excited states due to the state filling effect. It is found that the PL intensity of the first peak, which originates from the recombination of carriers at the ground state, is rapidly saturated when the excitation power is raised. In both samples, it is observed that the PL intensity increases initially with increasing excitation power. However, a reduction of the PL intensity occurs when the excitation power exceeds a critical value. This behavior is shown in Fig. 2. For the undoped QDs, the critical excitation power was found to be 250 mW. In comparison, a lower critical excitation power of 150 mW was observed for the p-doped QDs. It is generally believed that the reduction of PL intensity under high excitation powers is mainly caused by the increase of sample temperature. The photogenerated carriers that relax from high-energy states to low-energy ones will transfer their excessive energy to phonons, resulting in the heating of the lattices. The lower critical excitation power indicates that the temperature in the p-doped QDs rises more rapidly than that in the undoped QDs. It implies that the energy transfer rate from photogenerated carriers to phonons is larger in the p-doped QDs. In other words, it means that the relaxation of carriers is faster in

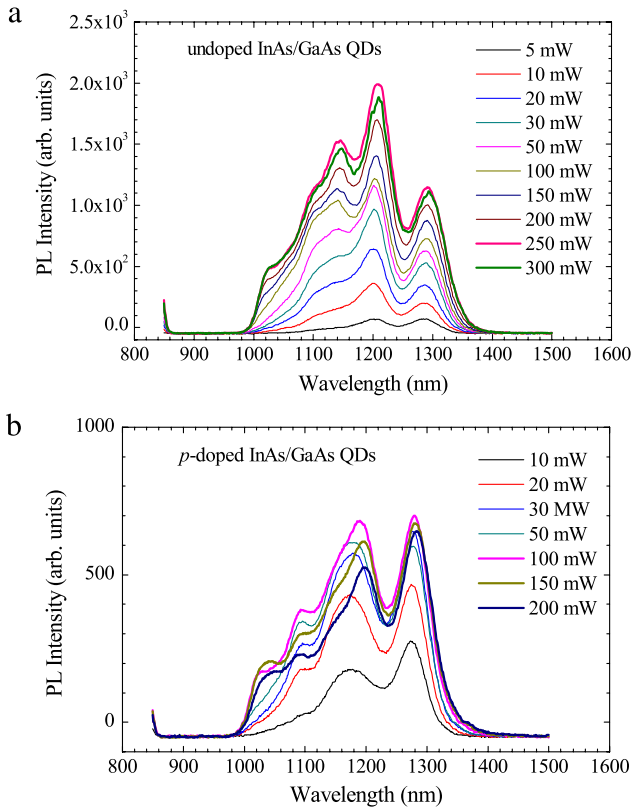

Fig. 2. Evolution of PL spectrum with increasing excitation power for (a) undoped and (b) p-doped InAs/GaAs QDs.

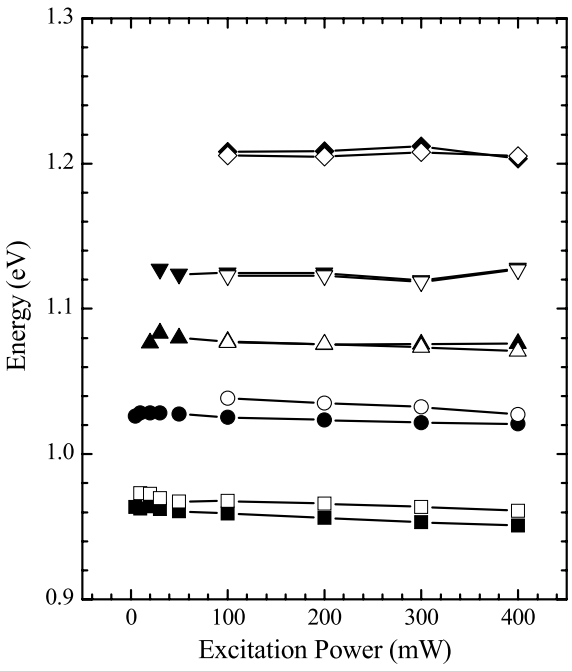

Fig. 3. Excitation power dependence of the transition energies for undoped (solid symbols) and p-doped (empty symbols) InAs/GaAs QDs.

the p-doped QDs due to the existence of a large number of build-in holes (about 10 holes per QD). This faster carrier relaxation under high excitation powers is also verified by pump–probe reflection measurements.

In order to determine the positions and linewidths for the discrete energy states of the QDs, the PL spectra of both samples obtained at different excitation powers have been decomposed by Gaussian fitting. The Gaussian fittings for the PL spectra of the two samples obtained at $P = 400$ mW are presented in Fig. 1. It can be seen that totally five emissions with Gaussian shapes are resolved at different positions under high excitation powers. Each emission corresponds to the energy transition between a pair of discrete energy states with the same quantum number for electrons and holes. Therefore, the decomposition of the PL spectra by Gaussian fitting allows us to extract the information of both the transition energy and linewidth for the discrete energy states of the QDs. The evolution of the transition energy with increasing excitation power is compared in Fig. 3 for the two QD samples. Apparently, the transition energy exhibits a linear dependence on excitation power and a red shift of the transition energy is observed with increasing excitation power. At high excitation powers, electrons and holes that have been captured into QDs may have chance to escape out of QDs through carrier–carrier scattering (i.e., Auger processes). These carriers may be recaptured into larger QDs with lower energies and eventually emit photons there through radiative recombination. It implies that more and more photogenerated carriers will be transferred to larger QDs and recombine there when carrier–carrier scattering becomes strong at high excitation powers. As a result, we observe a red shift of the transition energy with increasing excitation power. Apart from the red shift of the transition energy, it is also noticed that the transition energies for the p-doped QDs appear to be higher than those for the undoped QDs. With increasing excitation power, the difference in transition energy between the two QD samples remains nearly unchanged for the ground and first excited states while the difference in transition energy disappears for high-energy states. Physically, the major difference between the two QD samples is the large number of the build-in holes in the p-doped QDs (about 10 holes per QD). It has been known that the attractive or repulsive interaction between carriers would result in a decrease or an increase in energy states which is manifested in transition energy. At low excitation powers, the repulsive interaction between build-in holes in the p-doped QDs leads to an increase in the energy states of holes and thus in the transition energies. Consequently, the transition energies for the p-doped QDs appear at higher energy positions. At high excitation powers when the average number of photogenerated carriers per QD exceeds the build-in holes, the situations in the two QD samples become similar. As a result, the difference in transition energy disappears for high-energy states.

3.2. Effects of p-type doping on the carrier dynamics of QDs

Previously, we have performed pump–probe reflection measurements for the undoped and p-doped InAs/GaAs QDs samples at 800 nm under different pump powers and found out the influence of the injected carrier density on the carrier dynamics [20]. In order to clarify the influence of pump wavelength on carrier dynamics, we need to study the dependence of carrier dynamics on pump wavelength. The differential reflection spectra obtained at an excitation power of 120 mW and at different wavelengths for the undoped and p-doped InAs/GaAs QDs are present in Fig. 4(a) and (b), respectively. All spectra are normalized in order to make a clear comparison of the time constants for various processes. For both QD samples, one can see two fast decay processes with short time constants followed by a slow rise process with a much longer time constant in the differential reflection spectra. The three stages can be attributed to the transfer of photogenerated carriers from GaAs barrier into InAs wetting layer (characterized by a time constant τ_{b-w}), the capture of carriers from InAs wetting layer into InAs QDs (characterized by a time constant $\tau_{e,h}$), and the carrier relaxation and recombination inside InAs QDs. Obviously, the capture of carriers is much faster than the following relaxation and recombination of carriers. For the undoped QDs, it is noticed that for all excitation wavelengths the differential reflection becomes negative in

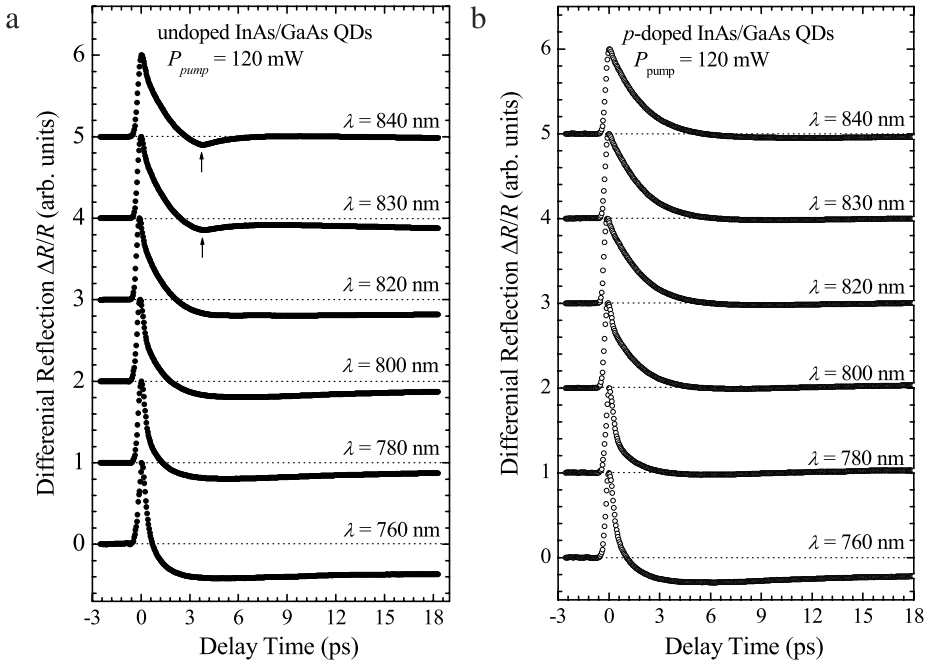

Fig. 4. Differential reflection spectra measured at different pump wavelengths for (a) undoped and (b) p-doped InAs/GaAs QDs.

the second stage when the photogenerated carriers are captured into InAs QDs. This is completely different from the previous report where the differential reflection remains to be a positive one in the decay process [19]. This phenomenon can be understood if we consider that the thickness of GaAs caper layer and InAs QD layer are different in our QD samples [20]. For the p-doped QDs, negative differential reflection appears only for excitation wavelengths shorter than 760 nm. For excitation wavelengths longer than 780 nm, we observe a significant slow down of the decay process because the reduction in the absorption of GaAs barrier with increasing excitation wavelength results in a smaller carrier density in GaAs barrier. Consequently, the capture of carriers, which may be facilitated by Auger processes, is slowed down. A remarkable feature observed in the p-doped QDs is the sudden appearance of the negative differential reflection when the excitation wavelength is slightly reduced from 780 to 760 nm. In this case, the change in the absorption of GaAs and thus the carrier density in it is not so large. However, this small change in the absorption results in a larger differential reflection in the second stage. Due to the large number of build-in holes, the low energy states of holes are already populated in the p-doped QDs prior to excitation. The electrons populating the lower energy states do not contribute to the change in the absorption of InAS QDs and thus the refractive index of QD layer and finally the reflection of the QD sample. Only when the high-energy states are populated by injected electrons, the absorption of the p-doped QDs would be modified. This situation can be realized by either increasing excitation density or decreasing excitation wavelength. Therefore, it is thought that the carrier density generated at ∼760 nm is large enough so that the injected electrons begin to occupy high-energy states. Since the negative differential reflection originates from the change in the absorption of InAs QDs after the capture of carriers from GaAs barrier into InAs QDs, this phenomenon implies that the change in the absorption of the undoped QDs is larger than that of the p-doped QDs due to the absence of build-in holes. For the undoped QDs, it is found that the negative differential reflection becomes smaller with increasing excitation wavelength. For excitation wavelengths longer than 820 nm, a rapid recovery of the differential reflection is observed, as indicated by arrows in Fig. 4(a).

Relying on the biexponential fitting of the differential reflection spectra, we are able to derive the time constants for the carrier capture and relaxation and recombination processes. In Fig. 5(a) and (b), we have plotted the time constants for the two capture processes (i.e., the two fast decay processes) as a function of pump wavelength for the undoped and p-doped QDs. The comparison is made only for excitation wavelengths shorter than 820 nm because of the appearance of the negative peak for excitation wavelengths longer than 820 nm. For the undoped QDs, τ_{b-w} and $\tau_{e,h}$ are found to be ∼0.90 and ∼1.85 ps at 815 nm. It implies that it takes ∼2.75 ps for photogenerated carriers to be captured from GaAs barrier into InAs QDs. With decreasing excitation wavelength, we observe a rapid decrease of τ_{b-w} to ∼0.30 ps at 790 nm and it remains nearly unchanged till 760 nm. On the other hand, the reduction of $\tau_{e,h}$ occurs at 805 nm and it decreases gradually to 0.90 ps at 760 nm. In the p-doped QDs, the value of τ_{b-w}, which is ∼0.30 ps, is almost independent of excitation wavelength. A gradual decrease of $\tau_{e,h}$ from 2.0 to 1.2 ps is observed in the wavelength range 785–800 nm. For excitation wavelengths shorter than 785 nm, $\tau_{e,h}$ does not change so much. It is generally thought that the shortening of capture time with decreasing excitation wavelength in the undoped and p-doped QDs is caused by increasing carrier density and enhanced Auger processes that may facilitate the capture and relaxation processes. Basically, the Auger scattering rate is proportional to the excitation density. As excitation wavelength decreases, the absorption coefficient of GaAs barrier becomes larger, leading to a higher carrier density and an enhanced Auger processes. For the p-doped QDs, the scattering of photogenerated carriers with the large build-in holes makes the transfer of carriers from GaAs barrier into InAs wetting layer fast. Consequently, the value of τ_{b-w} is not sensitive to excitation wavelength.

4. Conclusion

In summary, we have experimentally investigated the effect of *p*-doping on the optical properties of self-organized InAs/GaAs QDs by both μ-PL and degenerate pump–probe reflection measurements. The transition energies of the first five energy states have been determined by decomposing the PL spectra of QDs with

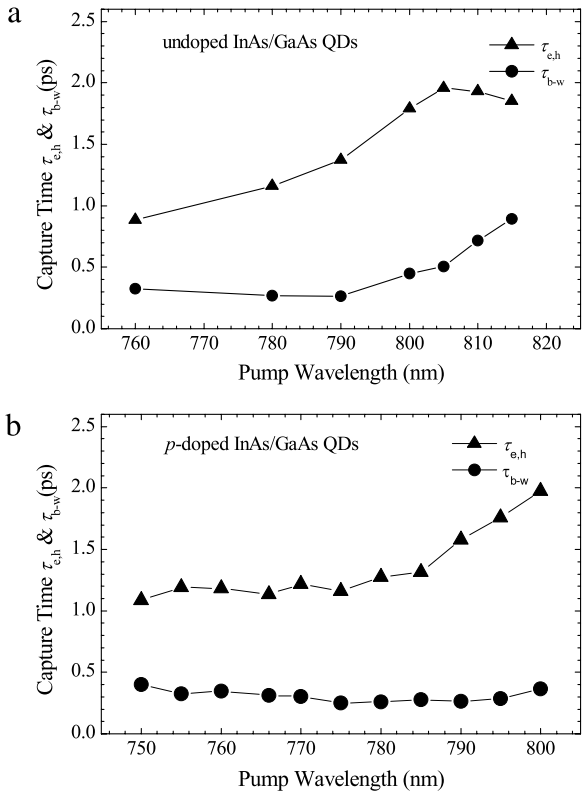

Fig. 5. Carrier capture and relaxation times derived at different pump wavelengths for (a) undoped and (b) p-doped InAs/GaAs QDs.

Gaussian fitting. A redshift of the transition energy and a broadening of the linewidth are observed with increasing excitation power. A reduction of PL intensity with increasing excitation power is observed when the excitation power exceeds a critical value. A lower critical excitation power is found for the p-doped QDs. The carrier dynamics in both QD samples has been investigated by degenerate pump–probe reflection measurements. The time constants for the capture and relaxation process have been derived and the excitation wavelength dependence of carrier dynamics is compared for the two QD samples. It is found that the large build-in holes in the p-doped QDs, which may facilitate carrier capture and relaxation processes, play an important role in determining carrier dynamics and it may be beneficial to the performance of QD-based devices.

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