Carrier dynamics and optical nonlinearity of alloyed CdSeTe quantum dots in glass matrix

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Abstract: Size and pump-fluence dependent ultrafast carrier dynamics of CdSeTe QDs are investigated using femtosecond pump-probe techniques operating at two different repetition rates: 1 kHz (low-repetition rate), and 76 MHz (high-repetition rate). With a low-repetition rate laser and 3.1 eV excitation photon energy, multiple exciton generation (MEG) is observed and the optical responses of alloyed QDs clearly show three components: a fast decay ascribed to carrier recombination, an intermediate component associated with MEG decay, and a slow decay associated with radiative Auger recombination. With a high-repetition rate laser and excitation photon energy resonant with band-edge energy, obvious coherent phonon oscillations are observed in 4 nm CdSeTe QDs due to impulsive stimulated Raman scattering. Open-aperture Z-scan measurement is used to clarify the size and pump-fluence dependence of optical nonlinearity under femtosecond laser excitation. With increasing laser power, an evolution from saturable absorption to reverse saturable absorption in CdSeTe QDs is observed. The transition process is analyzed using a phenomenological model based on nonlinear absorption coefficient and saturation intensity. These results indicate that CdSeTe QDs in a glass matrix are a class of materials for potential application in all-optical switching devices.

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1. Introduction

Semiconductor quantum dots (QDs) with three dimensions close to the Bohr radius of electron-hole pairs have received tremendous attention in the past several decades, owing to their unique physical and chemical properties. The quantum confinement and dielectric confinement effects make semiconductor QDs a promising class of materials for wide range of applications, such as solar cell [1], laser amplification [2], optical logic gate [3] etc. Novel photovoltaic technology based on multilayered nanofilms is one of the areas where QDs have been used to enhance efficiency through mechanisms like multiple exciton generation (MEG), phonon bottleneck and hot-electron transfer. PbSe, PbS and PbTe QDs as a series of narrow-band nanomaterials have been used to synthesize multilayered solar cells with MEG [4], but QDs containing poisonous lead are not preferred. Therefore, much emphasis has been put to alloyed II-VI QDs without poisonous elements, whose bandgap can be finely tuned within a broad range [5]. Glass is a transparent matrix for nanoparticles in the range of visible to infrared region. II-IV QDs with diameters of 1~20 nm have shown interesting optical and electronic properties due to the dielectric and electronic confinement [6, 7]. When the radius of a QD approaches the exciton Bohr radius, its electronic structure remarkably differentiate from its bulk counterpart because the oscillator strength of electrons is concentrated into a few discrete transitions [8]. However, so far few reports have been given about the size and pump-fluence dependent of carrier dynamics and optical nonlinearity for CdSeTe QDs in glass matrix.

Carrier dynamics and nonlinear absorption can provide comprehensive information to understand the mechanism of QDs interaction with strong laser pulse. Investigation on carrier relaxation dynamics in semiconductor QDs is of great importance from both the basic physics and application point of views. Photoexcited carrier dynamics in CdSe [9] and CdTe QDs [10] have been investigated by several groups on picosecond and femtosecond time scales. Chai et al. [11] reported coherent optical phonons in CdSeTe QDs embedded in a glass matrix using high-repetition-rate femtosecond laser. The amplitude of the observed optical phonons exhibits a resonance that can be well described by a model based on impulsive stimulated Raman scattering. B.T. Spann identified the Raman active CdSe and CdTe longitudinal optical modes in CdSeTe QDs using femtosecond transient transmission spectroscopy operating at a repetition rate of 1 kHz, and quantified the strength of electron-phonon coupling [12]. F. J. Zhang reported higher order recombination in CdSSe QDs-doped glass matrix and the origins have been attributed to broad size, state distribution and high-pump power. Auger recombination has also been shown to play a key role in the multi-exciton recombination [13]. Carriers are firstly pumped from the ground state to the excited states, followed by broadband absorption of these excited carriers due to the existence of high-lying states or continuum states. Then the excited carriers relax to the ground state through electron-electron coupling, electron phonon interaction and phonon-phonon relaxation [14]. Recently, several research groups [15, 16] have begun to investigate the mechanism of carrier dynamics of alloyed CdSeTe QDs for higher solar cell efficiency. However, there is still a lack of systematic study its dynamics and power-dependence behavior. Combined studies of size and pump-fluence dependence of ultrafast dynamics and optical nonlinearity for alloyed CdSeTe QDs are appropriate to understand their properties and to develop devices with high performance.

In this paper, the size and pump-fluence dependence of carrier dynamics and nonlinear properties of three types of CdSeTe QDs are investigated using pump-probe transmission and Z-scan measurement, respectively. The samples under study have almost the same chemical composition, and only change in the average QDs size. The carrier lifetimes are investigated as a function of pump fluences. Z-scan measurements reveal both saturable absorption and reverse saturable absorption. With the pump fluence increasing, the evolution from saturable absorption to reverse saturable absorption is observed.
2. Experimental technique

2.1 Preparation of samples and basic optical characterization

In order to eliminate the influence from the ligands or solvents on carrier dynamics, we choose three sizes of CdSeTe QDs, embedded into the melted borosilicate glass at about 1300°C in a gradient furnace. The composition of borosilicate glass can be found in the work of Banfi et al [17]. The bandgap for bulk CdSe and CdTe is around 1.74 eV (712 nm) and 1.50 eV (827 nm) at room temperature, with exciton Bohr radius 5.6 nm and 7.4 nm, respectively [18]. The compositions of QDs are CdSe$_{0.32}$Te$_{0.68}$ (3.9 nm radius, 1.59 eV bandgap), CdSe$_{0.35}$Te$_{0.65}$ (5.3 nm radius, 1.49 eV bandgap) and CdSe$_{0.32}$Te$_{0.68}$ (11.0 nm radius, 1.46 eV bandgap); and we name them as 4 nm, 5 nm and 11 nm, respectively.

The samples are characterized by both absorption and Raman spectroscopies, with confocal DXR Raman system using a 514 nm laser with 2 mW power. As shown in Fig. 1(a), due to quantum confinement, with increasing QD size, absorption spectra of three samples show absorption edges near 755, 823 and 837 nm, respectively, consistent with the bandgap values. As shown in Fig. 1(b), main Raman peaks are at 199, 196 and 189 cm$^{-1}$ for 4 nm, 5 nm and 11 nm, respectively, showing that phonon frequencies decrease with increasing QD size. For 4 nm sample, two clear peaks are attributed to longitudinal optical modes with 199 cm$^{-1}$ for the CdTe-like phonon and 156 cm$^{-1}$ for the CdSe-like phonon, both of which have been investigated in previous study using single-color pump-probe spectroscopy [11].

![Fig. 1. (a) Absorption spectra of 4 nm, 5 nm, 11 nm filter near the absorption edge. The absorption peak can be determined by taking the derivative of absorption spectra. (b) Raman spectra of 4 nm, 5 nm, 11 nm QD samples using 514 nm laser excitation.](image)

2.2 Femtosecond pump-probe and Z-scan experiment

The ultrafast dynamics of QDs are studied with femtosecond laser pump-probe spectroscopies using two different high-repetition-rate and low-repetition-rate Ti: Sapphire femtosecond laser systems. Details about the high-repetition rate (76 MHz) pump-probe spectrometer can be found in Ref [11]. Experiments with low-repetition rate (1 kHz) pump-probe spectrometer are performed in collinear transmission geometry at room temperature. Laser pulses generated from Ti: sapphire regenerative amplifier (Coherent, Legend, 1 kHz) have 120 fs pulse width (FWHM) and 800 nm central wavelength. A second-harmonic generation crystal (BBO) is used to double the photon energy to 400 nm (3.10 eV), after which a band-pass filter is used to block the residual laser at 800 nm (1.55 eV). The collinear pump at 3.1 eV and probe at 1.55 eV are focused onto the sample surface by a 150 mm lens with the spot sizes of 80 and 50 µm (diameters), respectively. The transmitted laser beam is detected with a balanced Si detector (Model 2307, Newport) and then amplified by a preamplifier (Model SR560,
Stanford Research Systems). A reference beam split from the probe beam is also directed into
the balanced detector to improve the signal-to-noise ratio. Neutral density filters are used
before the detector to ensure that the detector operates in the linear regime. Lock-in amplifier,
which works with a mechanical chopper running at 85 Hz in pump beam path, is used to
collect data. The thickness of the samples is 2 mm, and the cleaved surface is perpendicular to
the laser incident direction.

The open-aperture Z-scan measurements are carried out with femtosecond laser pulses
(Coherent, Legend) operating at 800 nm, 1 kHz to avoid thermal accumulation. The laser
beam is focused by a 150 mm lens. The focused beam waist is estimated to be 20 µm, and the
Rayleigh range \( z_0 \) is about 1.6 mm, which satisfies the thin medium conditions [19]. The
incident and transmitted laser powers are monitored as the samples move along the laser
propagation direction. The open-aperture Z-scan signals are collected with a Silicon detector
(Thorlabs, Det36A) and then recorded by a lock-in amplifier (EG&G, Model 5210). The pulse
energy is measured by an energy meter of Quantum Energy Max sensors (Coherent, J-10SI-
HE).

3. Results and discussion

3.1. Carrier dynamics of CdSeTe QDs under high- and low-repetition rate laser
excitations

As shown in Fig. 2(a) and 2(b), time-resolved absorption measurements have been conducted
in all three QD samples, under both high-repetition rate (76 MHz, 800 nm pump, 800 nm
probe) and low-repetition rate (1 kHz, 400 nm pump, 800 nm probe) excitations. All
absorption signals show sharp increase due to electron excitation, then decay at various time
scales due to electron-electron (e-e) and electron-phonon (e-ph) coupling. The optical
transmission responses of all three QD samples show three regions: a sharp increase due to
carrier excitation, fast decay corresponding to carrier recombination, and slow decays because
of thermal response. Coherent phonon oscillations are only observed in the 4 nm sample
under high-repetition rate excitation at 1.55 eV, of which the photon energy resonates with
QD bandgap. The carrier contribution can be described with a set of decaying exponential
functions, representing different electron relaxation mechanisms that vary in specific samples
under different pump-probe conditions. The coherent phonon oscillations can be described
with a set of damping oscillators, with damping rate and oscillation frequency specified for
each phonon mode. An analytical expression consisting of multiple exponential and damping
oscillations convoluted with instrumental response [20] is used to fit our transient absorption
signals:

\[
-\frac{\Delta A}{A}(t) = \sum_i a_i e^{-(t-t_i)/\tau_i} + \sum_l \beta_l e^{-(t-t_l)/\gamma_l} \cos(\omega t + \phi_l) H(t)
\]

where \( a_i \) and \( \tau_i \) are the amplitude and lifetime of the \( i \)th term in electron relaxation processes,
respectively. \( \beta_l \) and \( \gamma_l \) represent the amplitude and lifetime of the \( l \)th term in coherent phonon.
\( H(t) \) is the Heaviside function convoluted with Gaussian to account for the instrumental
response time \( \tau_{FWHM} \) of pump and probe pulses. Carrier dynamics of CdSeTe QDs depends on
the absorbed laser energy, size, excitation wavelength and their surrounding matrix. Several
mechanisms, such as Coulomb blockade effect [21] and Auger recombination [22] have been
proposed to explain pump-fluence dependence of carrier dynamics in II-VI QDs. In order to
investigate the transition mechanism, the number of photons absorbed per QDs \( N_{abs} \) is
calculated as [4]

\[
N_{abs} = J(0)(1-e^{-\alpha L})/cL
\]

where \( L \) is the thickness of the sample, \( J(0) \) is the photon flux on the sample’s surface, \( \alpha \) is the
absorption coefficient at the pump wavelength, and \( c \) is the concentration of QDs in the glass
matrix. We estimate \( N_{abs} \) is 3.51 when pump fluence is 1 mJ/cm^2 at 3.1 eV, multiple exciton
generation (MEG) exists under low-repetition rate laser excitation at 3.1 eV, 10 μJ/cm² at 1.55 eV is corresponding to $N_{ph}$ of 0.0008 for 11 nm QDs sample.

With low-repetition rate laser excitation, photon energy of pump laser (3.10 eV) is about twice of the bandgap in all three samples, hence MEG can occur after pump photons are absorbed. No coherent phonon of CdSeTe QDs is observed in any sample. As shown in Fig. 2(b) and 2(d), the time-resolved absorption change for all three samples at various pump fluences can be fitted by three exponential decays, with a fast decaying component $\tau_1$, an intermediate component $\tau_2$ and a slow decaying component $\tau_3$. The fast decaying component $\tau_1$ is ascribed to the relaxation associated with carrier recombination. It is found that $\tau_1$ decreases with the increasing pump fluence, and the reason is that high pump fluence leads to strong absorption and higher concentrations of carriers, which shortens the tunneling distances for recombination and increases the probability of recombination [23]. The intermediate component $\tau_2$ is associated with MEG decay (non-radiative Auger recombination) [24]. MEG is similar to Auger recombination. For Auger recombination an electron and a hole recombin, and the released energy is transferred to another charge carrier, which leads to the excitation of a third electron or hole [25]. For MEG, when one exciton recombines, it can transfer energy to another exciton and cool down rapidly through exciton-exciton and exciton-phonon interactions. Both MEG and Auger recombination require high concentration of excited carriers, which can decrease the tunneling distances for recombination and increase the probability of electron-hole recombination. MEG also requires that the rate of multi-exciton recombination is much faster than single exciton recombination [26]. The slow decaying component $\tau_3$ is assigned to radiative Auger recombination, which is not sensitive to pump fluence [27].

Under high-repetition rate laser excitation, in 5 nm and 11 nm samples, the signals can still be fitted by three exponential decays, with a fast decaying component $\tau_1$, an intermediate component $\tau_2$ and a slow decaying component $\tau_3$. Comparing with low-repetition-rate cases, $\tau_1$ and $\tau_2$ have similar values and trends, but $\tau_3$ is about 5 times longer, which is a result of thermal accumulation. The thermal accumulation effect will show up when the time interval between incident laser pulses is much shorter than the characteristic thermal time constant (ms scale for glass) [28]. Local temperature around QDs will increase until a steady state is reached when the heat generation rate equals to heat dissipation rate through conduction. The repetition rate of our laser is 76 MHz, so the time interval between two pulses is 13.1 ns. Therefore, for 5 nm and 11 nm, the long component ($\tau_3$) under the high-repetition rate laser excitation, is much longer than the low-repetition rate case. Comparing with the 5 nm and 11 nm samples, carrier dynamics of 4 nm sample is quite different. In the 4 nm sample, the signals are fitted with double exponential decay and two damping harmonic oscillators, latter of which reflect the excited coherent phonons. There is no slow decay component $\tau_3$ for 4 nm. Also the decay times $\tau_1$ and $\tau_2$ of 4 nm are much shorter than those of 5 nm and 11 nm, with weak dependence on pump fluences.

Exciton generation creates a static electric field that can cause the bandgap reduction [29]. For 4 nm sample, the 1.59 eV bandgap is close to the probe photon energy (1.55 eV). Excitation with higher pump fluence can lead to carrier induced Stark effect (CISE), from which more probe photons can be absorbed [30]. Coherent phonon can be generated due to this resonant absorption. Due to the energy transfer into coherent phonon lifetime $\tau_1$ and $\tau_2$ become much shorter, only down to several picoseconds. In other words, the efficiency of coherent phonon generation is enhanced when the photon energy matches the electronic transition energy. For 5 nm and 11 nm samples, excitation with higher pump fluence generates more excitons, which decrease the tunneling distance for recombination and increase the probability of electron-hole recombination [31]. With increasing pump fluence, the lifetime of exciton generation ($\tau_2$) and Auger recombination ($\tau_3$) become shorter, but thermal accumulation time ($\tau_3$) becomes longer. For 4 nm sample the number of particles at excited states is less than those of 5 nm and 11 nm samples, of which relaxation lifetimes are very short down to several picoseconds. It is easy to absorb enough photon to another higher excited state. Because there are stronger absorption in 5 nm and 11 nm samples, the excited
state is probably saturated with electrons due to strong absorption in 5 nm and 11 nm samples. So we can see longer lifetime in 5 nm and 11 nm samples.

The disappearance of slow decay component in 4 nm sample under high-repetition rate excitation could relate to the generation of coherent phonons, which provides a fast energy relaxation channel for excited carriers. In 4 nm sample, under resonant laser excitation, coherent phonons exist because of Fröhlich coupling, and can be described by a model based on impulsive stimulated Raman scattering [32]. For 5 nm and 11 nm samples, under off-resonant laser excitation, carriers in QDs relax to ground state directly with a long lifetime. Trapping states in the bandgap can also work as an intermediate state for the transfer of charge carriers. The term with long lifetime \( \tau_3 \) (~100 ps) under off-resonant excitation originates from thermal accumulation, which cannot diffuse to surrounding medium.

Figure 3(a) shows the coherent phonon oscillations of 4 nm sample at pump fluence of 44 \( \mu J/cm^2 \), from which the non-oscillatory components have been removed by applying a digital band-pass filter. Figure 3(b) plots the Fast Fourier Transform of phonon oscillations, and two frequency components have been revealed. The higher frequency component at 6.04 THz is CdSe-like phonon mode, while the lower frequency component at 4.83 THz is CdTe-like phonon mode. FFT results of phonon oscillations agree with Raman shifts shown as black dots. The slight difference between Raman and coherent phonon frequencies comes from the fact that excited states near electronic resonance energy are probed in pump-probe experiment, while Raman shift probes the ground state. The peak shift compared with steady-state Raman spectrum is consistent with the fact that QDs are excited resonantly in 4 nm sample. Figure 3(c) and 3(d) present the change of phonon amplitude and phonon lifetime for each mode at various pump fluences. With higher pump fluence, the amplitude of CdSe-like
phonon mode becomes larger, and its lifetime becomes longer, from ~0.6 ps to ~1.5 ps due to heat accumulation between pulses [33].

Comparing the data taken under low-repetition and high-repetition rate excitations, it can give insights of carrier dynamics in CdSeTe QDs. Single exciton and MEG for CdSeTe QDs possess excitation frequencies on the same order. We can see \( \tau \) but under the resonant excitation condition, coherent phonon is observed and the signal can be fitting with two decay terms, which are much smaller than another two samples.

Fig. 3. (a) Coherent phonon vibrations and fitting in 4 nm sample under pump fluence of 44 \( \mu \)J/cm\(^2\). (b) Fast Fourier Transformation of coherent phonon signals as a function of the pump fluence. c) and d) Change of amplitude and lifetime of coherent phonons under different pump fluences.

3.2 The nonlinear optical property of CdSeTe QDs

In order to understand the size- and fluence-dependence of nonlinear absorption in CdSeTe QDs and analyze the influence from resonant and off-resonant excitation, open-aperture Z-scan measurements are performed to reveal the absorption mechanisms. In order to validate our femtosecond open-aperture Z-scan experiment, we firstly measure the nonlinear absorption coefficients of carbon disulfide. We got \( \beta = 1.5 \times 10^{-2} \text{ cm/GW} \) at pump density of \( I_0 = 200 \text{ GW/cm}^2 \), which is consistent with the reported results [34]. The open-aperture Z-scan measurements are performed with 1.55 eV photon energy under low-repetition rate excitation. With low-repetition-rate laser pulses, thermal effect can be safely neglected. Results for all the three samples at different fluences are presented in Fig. 4. Generally speaking, optical response of nanoparticles consists of both absorption and scattering, the latter can play a significant role for relatively large nanoparticles [35]. In order to utilize theoretical models to analyze the nonlinear absorption process, a more-general situation is considered here, including both saturation intensity \( I_s \) and absorption coefficient \( \beta \). We combine the effects of saturable absorption (SA) and reverse saturable absorption (RSA), and then yield the total absorption coefficient as:
where $\alpha_0$ is the linear absorption coefficient which is $1.49, 1.02 \times 10$ and $1.81 \times 10$ cm$^{-1}$ for 4 nm, 5 nm and 11 nm at 1.55 eV, respectively, as derived from Fig. 1(a). $I$ and $I_s$ are laser intensity and saturation intensity, respectively. $\beta$ is the positive nonlinear absorption coefficient. The first term represents saturable absorption $\alpha_{sa}(I)$ and the second term represents reverse saturable absorption $\alpha_{rsa}(I)$. Then the normalized transmission can be expressed as [36]:

$$T(z) = \sum_{m=0}^{\infty} \left( \frac{-\alpha(I)I_0L_{eff}}{I(1+z^2/z_m^2)} \right)^m \frac{1}{(1+m)^{1/2}}$$  (4)

where $L_{eff}$ is the effective optical path, and $z$ is the displacement of sample from the focal point. It can be seen from Fig. 4 that the theoretical fitting for all three samples is in good agreement with the experimental data. The estimated values of saturation intensity and nonlinear absorption coefficient $\beta$ are given in Table 1. From Fig. 4, we can see the evolution of signal shape of open-aperture Z-scan of CdSeTe QDs in glass matrix under different laser fluences. When the sample is far from the focus, the linear absorption dominates, without any nonlinearity observed, hence the transmittance is unity. As the sample moves towards the focus, the moderate intensity induces increase of the population of the carriers due to nonlinear absorption.

For 4 nm sample, the reverse absorption (RA) occurs under higher ultrafast laser excitations at 6.0 GW/cm$^2$, because the bandgap of 4 nm sample is 1.59 eV, larger than laser photon energy of 1.55 eV, so two-photon absorption (TPA) is observed. Under lower laser excitation, no obvious RSA can be observed. From Table 1, we can see nonlinear absorption coefficient $\beta$ of 4 nm sample increases with laser intensity.

For 5 nm and 11 nm samples, under low laser intensities, the saturation absorption happens because their energy bandgaps are smaller than laser photon energy. At low laser intensity, as the sample moves towards the focus, electrons at valence band are excited to the conduction band, thus the majority of the nanoparticles are pumped to the excited state, leading to a smaller population at the ground state. At this intensity level, the transmitted intensity will be at maximum which results in saturation absorption (SA) behavior [37, 38]. Carriers are firstly pumped from ground state to the excited state where they then become free carriers, this process is subsequently followed by the broad band absorption of these free carriers. Then the excited electrons relax to the ground state by electron-electron coupling, electron-phonon interaction and phonon-phonon relaxation. At low pump fluence, open-aperture Z-scan shows saturable absorption. When the sample is far from focal point, transmission through the sample is low due to strong linear absorption. As the intensity increases near the focal point, the population of electrons saturates the conduction band, thus blocks further excitation from valance band, hence, transmission through the sample increases. This is one of the phenomena that is responsible for passive mode locking in lasers. Such reverse saturation and saturation absorption makes CdSeTe QDs an ideal candidate for optical limiting and all-optical logic gate applications. The majority of the QDs are pumped to the excited state, leading to a smaller population at the ground state, this causes the bleaching in the ground-state absorption band. Furthermore, when pump intensities increase to tens of GW/cm$^2$, the combination of SA and RSA with opposite signs is observed [39]. The strong excited-state carrier absorption result in an obvious dip in the transmission signals near the focal point. With higher laser intensity, the dip drags the transmission signals towards unity. These results indicate that RSA only occurs at extremely high carrier intensities, but its effect may totally offset the SA effect with high enough laser intensity.
Fig. 4. The open-aperture (S = 1) Z-scan curves for CdSeTe QDs (4 nm, 5 nm, and 11 nm) at 1.55 eV femtosecond excitation. The solid lines are the theoretical fitting curves.

<table>
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<th>Pump fluence</th>
<th>( \beta ) cm/GW</th>
<th>( I_0 ) GW/cm²</th>
<th>( \beta ) cm/GW</th>
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<tr>
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<td>8.0</td>
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5. Conclusions

In summary, three alloyed CdSe\(_{1-x}\)Te\(_x\) QDs with radii of 4, 5 and 11 nm are fabricated by melting-process method in the glass matrix and their carrier dynamics and optical nonlinearity are measured with pump-probe and Z-scan experiments. Under high-repetition rate excitation, coherent phonon oscillations are generated via Fröhlich coupling, but only observed in 4nm sample, where the photon energy is resonant with band-edge energy, and longer lifetime of carriers for CdSe\(_{1-x}\)Te\(_x\) QDs (R = 5, 11 nm) is observed. In samples 5nm and 11nm, with photon energy off resonant with band edge, carriers in QDs relax to ground state directly with much longer lifetimes. Under low-repetition rate excitation, multiple exciton generation (MEG) is responsible for the observed carrier dynamics. Z-scan measurements indicate that saturable absorption and reverse saturable absorption. With increasing pump fluence, the evolution from saturable absorption to reverse saturable absorption is revealed and nonlinear absorption coefficients have been obtained.

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