Electronic Supplementary Material (ESI) for Nanoscale.
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Supplementary information

Figure S1 shows the experimentally measured pump and probe beam size. By carefully scanning the knife-edge and then fitting to an integrated Gaussian profile, we can obtain the accurate pump and probe beam size of $38 \pm 0.6\,\mu m$ and $20.5 \pm 0.5\,\mu m$, respectively. By using these two beam sizes, we can convert the pump power to the absorbed fluences and then calculate photogenerated exciton density. The fitting model for obtaining the beam size is as the following:

$$y_x = y_0 + \int_{-\infty}^{+\infty} \frac{A}{\omega\sqrt{\pi/2}} \exp(-\frac{2(x-x_c)^2}{\omega^2})dx$$

where the ω is the full width at half maximum (FWHM) of the beam profile.

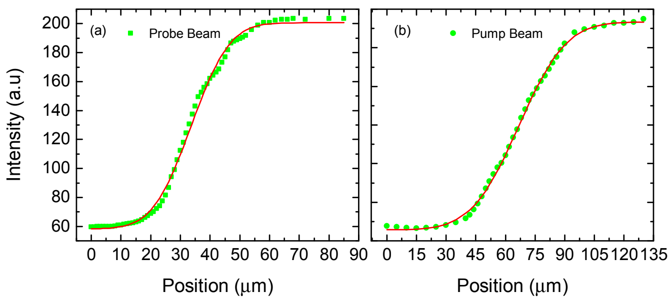

Figure S1 The measured beam size of the probe pulse (a) and the pump pulse (b). The vertical axis shows the reflected pulse intensity by the scanning knife-edge and the horizontal axis is the position of the moving knife-edge. The red solid lines represent the fitting line obtained by the integration of Gaussian function.

Fig.S2 shows the fitting results of Eq. 1without the constant term. We fitted the data between $\Delta T = 5.6\,ps$ and $169\,ps$ to the Eq. 1 (shown in the main text). Although the experimental data can be well reproduced by the Eq. 1 , the obtained fitting parameter γ is found to be increase with the decrease of the pump fluences below $1.62\,\mu J/cm^2$. This result indicated that below $1.62\,\mu J/cm^2$, additional relaxation channel is required for the excitons. Figure S3 presents the photogenerated excitons relaxation dynamics that corresponds to the component. It is obvious in the Figure. S3 that at the left part of the vertical solid line, the $\Delta R/R$ traces under different pump fluences are almost the same indicating the remained population of excitons is approximately equal after the initial and relaxation process. Based on this observation, we conclude that the decay component which is assigned to be the electron-hole recombination of isolated excitons should be independent of exciton density while the additional decrease trend arising from the contribution after $\Delta T = 400\,ps$ may be due to the accumulated heat effect.

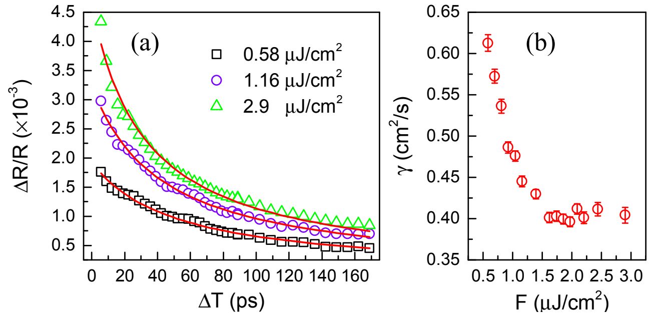

Figure S2 (a) The same experimental data as shown in Figure.3 (a) but fitted by the Eq.1 with different annihilation rate γ. Different open symbols here represent the experimental data under different pump fluences and the red solid lines are the fitting results. (b) Fitted parameter γ of exciton annihilation rate as a function of pump fluences without adding the constant term.

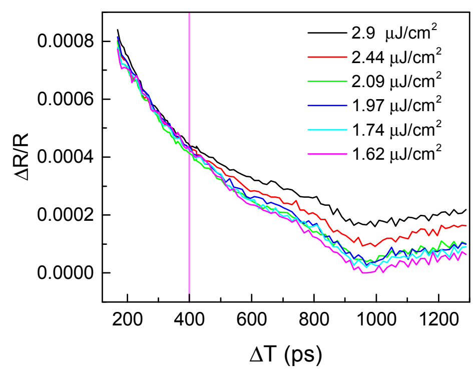

Figure S3 Transient reflectivity spectra at different pump fluences between $\Delta T = 200 - 1300\,ps$ which corresponding to the decay component of τ_3. The vertical line divides the curves into two areas at $\Delta T = 400\,ps$.