Applied Physics Letters

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Cite as: Appl. Phys. Lett. **119**, 192103 (2021); https://doi.org/10.1063/5.0073014
Submitted: 27 September 2021 • Accepted: 25 October 2021 • Published Online: 11 November 2021

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Applied Physics Letters ARTICLE scitation.org/journal/apl

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ABSTRACT

We hetero-paired a ∼200 nm thin film consisting of colloidal 2.5 nm PbS quantum dots deposited on semi-insulating GaAs. By exciting the thin film with laser pulses (26 ps, 10 Hz) at 1064 nm, we observed the two-photon stimulated emission of the PbS quantum dots and the GaAs host. At a certain intensity of the optical stimulus, the absorption capability of the quantum dots collectively saturates, and more photons of the laser beam reach the GaAs host, causing a bistable-like up-switch in the GaAs photoluminescence intensity. The work further addresses the determination of the two-photon absorption coefficient, which was found to be 8.6×10^{-6} m/W.

Semiconductor quantum dots (QDs) have always enjoyed a great deal of attention due to their resemblance to artificial atoms and their applicability as an active medium in optoelectronic devices. Lead sulfide (PbS) QDs are particularly appealing platforms to explore various properties of quantized semiconducting matter and, therefore, continue to attract ongoing research. The investigated properties in the above material systems include dielectric function,[1] photoluminescence (PL),[2–6] transmission,[4,7] thermally modified bandgap (E_g) variations,[4,7,8] magneto-optical effects,[9,10] and doping, either achieved chemically[11] or photo-dynamically, among other parameters.[12,13] On the applied research front, hetero-pairings between PbS QDs and numerous materials, such as oxides,[11] GaAs,[14–18] polyethylene terephthalate (PET),[19] and graphene,[20] have shown possibilities for device realizations and their integration into existing technologies. Potential applications include solar cells,[11,21] futuristic optically activated pn-junctions,[13,17,22] chromatically stable PL sources due to inherent lattice cooling,[7,16] non-brittle nano-electronics,[19] near-infrared photodetectors,[20,23] and less invasive medical diagnostics using moderate magnetic fields.[9,10,22]

In this work, we focus on the "secondary effects" of QDs, i.e., their influence on the hetero-paired partners, such as GaAs. We point out that the presence of PbS QDs alters E_g,[16] Urbach tail steepness,[16,17] and considerably the PL properties of undoped GaAs wafers.[18] As a consequence of our former work reported on two-photon excited PL (TPL), previously reported in Ref. 13, and further expanded in Ref. 18, we present herein an application of PbS QDs, namely, an all-optical switching device.

As described in more detail elsewhere,[9,10,18] approximately 200 nm thick QD films were deposited on semi-insulating GaAs substrates by a simple drop-casting technique. The employed colloidal oleic acid capped QDs have a diameter of 2.5 ± 0.3 nm ($E_g \sim 1.25$ eV) and were excited with the 1064 nm (1.17 eV) emission of a pulsed Nd:YAG laser (26 ps, 10 Hz). Since the laser's emission energy is smaller than E_g of the QDs, the irradiation caused TPL. The PL emissions were collected at room temperature (RT) in reflection geometry employing a fiber spectrometer.

Figure 1 shows selected spectra of the PbS QD/GaAs sample. The PL peaks of GaAs and PbS QDs are distinctively visible; however, a laser pulse intensity exceeding ∼55 MW/cm^2 was required to cause GaAs PL. Visualized by square and round symbols, the comparison of the integrated peak intensity (I_{TPL}) vs excitation intensity (I_{ex}) for PbS QDs and GaAs is shown in Fig. 2. We note that the uncertainty of the data closely coincides with the symbol size used. The dotted and broken lines are fits of the trends. The fits were performed with[18]

$$I_{TPL} \propto \left[\left\{ 1 + 4\sigma I_{ex} \right\}^{1/2} - 1 \right], \tag{1}$$

and

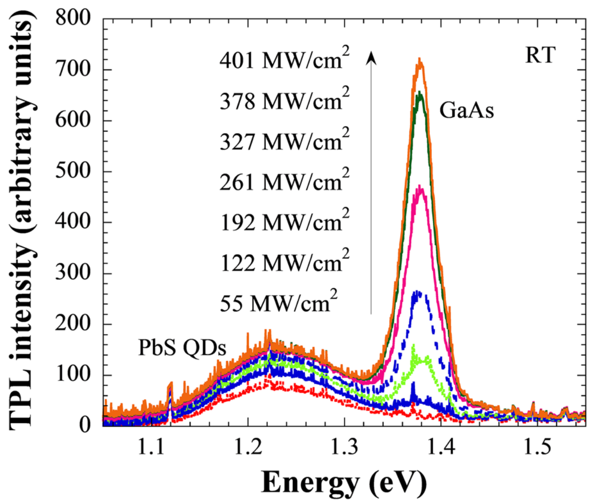

FIG. 1. Selected TPL spectra of PbS QD/GaAs measured at various impinging laser pulse intensities. The emissions of the GaAs host and the PbS QDs are clearly visible.

$$I_{TPL} \propto I_{ex}^{c}, \tag{2}$$

for the PbS QDs and GaAs, respectively, where σ is the slope parameter (0.59 cm^2/MW), and $c = 2.8$. The validations of the fits are discussed in Refs. 6 and 18 and are not a particular subject of the current paper. We focus here on the unsteady sudden increase in the GaAs TPL at $I_{ex} \sim 225$ MW/cm^2 and notice that it coincides with the threshold of complete stagnation of the QD emission. The situation is visualized by arrows in Fig. 2, and, in order to emphasize the unsteadiness, by curves (a) and (b) in Figs. 3 and 4, we present the comparison of

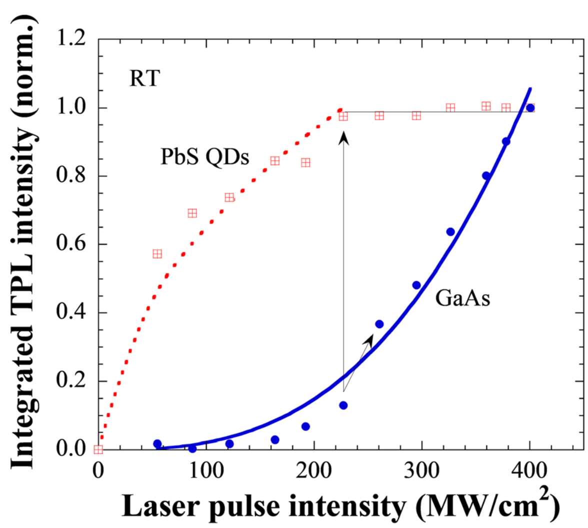

FIG. 2. The symbols show I_{TPL} of GaAs and PbS QDs vs laser pulse intensity of the spectra in Fig. 1, whereas the lines are fits described in the text. Indicated by arrows, the saturation of the PbS QD TPL, which is indicated by the horizontal line, coincides with a step-like increase in the GaAs TPL.

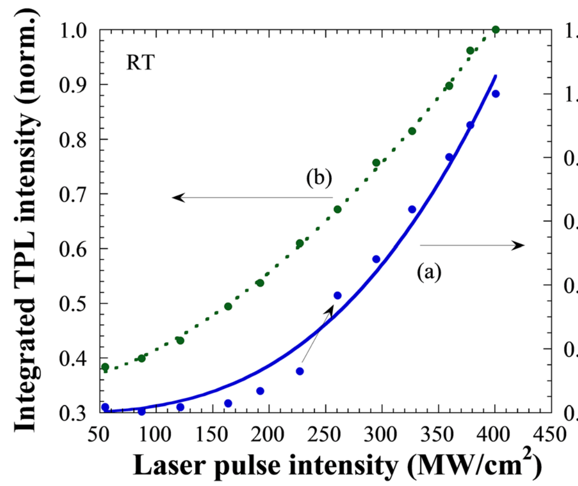

FIG. 3. (a) GaAs TPL in Fig. 2 in comparison with (b) GaAs TPL without a PbS QD film. The symbols represent I_{TPL}, and the lines are fits described in the text, and Ref. 18. Marked with the arrow, the PbS QD/GaAs TPL shows out of sequence a sudden increase at $I_{ex} \sim 225$ MW/cm^2, which is not present in the TPL trend of the GaAs wafer.

I_{TPL} in Fig. 2 to a GaAs substrate without QD cover, and the corresponding numerical derivatives, respectively.

The experimental setup can be used to determine the two-photon absorption coefficient β. Looking at Fig. 1, at $I_{ex} \sim 100$ MW/cm^2, the incoming laser light just starts to penetrate the QD thin-film provoking TPL of GaAs. With $d_p = 200$ nm using,[24]

$$\beta = \frac{(e-1)}{I_{ex} d_p}, \tag{3}$$

we find $\beta = 8.6 \times 10^{-6}$ m/W. This result differs from 1.4×10^{-9} m/W for films formed with 3.3 nm QDs embedded in a glue matrix.[25]

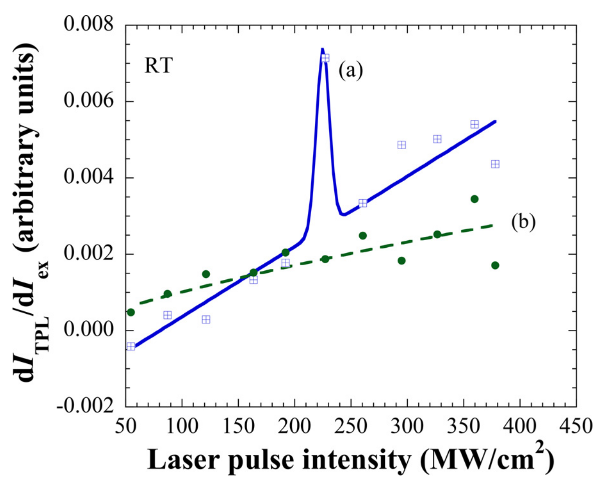

FIG. 4. Corresponding numerical derivatives of the I_{TPL} trends in Fig. 3 for (a) PbS QD/GaAs, and (b) stand-alone GaAs. The comparison clearly shows the switch and reveals that the derivative of the PbS QD/GaAs sample exhibits clearly more variation than the one of GaAs.

Possibly, the embedding host and the impinging laser excitation (120 fs pulses in Ref. 25, resulting in intensities of $\sim$1 GW/cm^2) influences β.

Summarizing, based on Figs. 3 and 4, we interpret the observed effect as an all-optical switch acting as an intensity filter, which starts to become more transmissive at a certain impinging light intensity. Our conclusion is based on Ref. 6 in which we pointed out that the excitation of one electron–hole pair per QD defines the PL saturation threshold of a QD ensemble. It is the case beyond $I_{ex} \sim 225$ MW/cm^2, and therefore, no transitions in the QDs are left to be excited. Consequently, the QD film is more transparent, and more photons impinge the GaAs host producing a more intense GaAs emission. Notably, the saturation effect manifests itself as a bistable property within the PbS QD film, leading to a collective optical switch for the transmitted emission of the GaAs host. Deduced from the TPL spectra in Fig. 1, we pointed out that $\beta = 8.6 \times 10^{-6}$ m/W for oleic acid capped colloidal 2.5 nm PbS QDs.

This work was partially supported by Miami University Regionals CLAAS Research Fund (Spring 2021).

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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