



# A New Spatiotemporal Scanning Technique for Two-Photon Fluorescence

Yizhi Zhu , Qiannan Cui , \* and Chunxiang Xu ,\*

State Key Laboratory of Bioelectronics, School of Biological Science and Medical Engineering,  
Southeast University, Nanjing 210096, China

\*Corresponding author: [qiannan@seu.edu.cn](mailto:qiannan@seu.edu.cn) ; [xcxseu@seu.edu.cn](mailto:xcxseu@seu.edu.cn)

Two-photon laser scanning microscope (TPLSM) provides outstanding optical three dimension section properties and it has been widely used in fundamental science and biomedical application. However, Current 3D two-photon fluorescence (TPF) imaging techniques usually overlook the spatiotemporal evolution of TPF ellipsoid along the axial direction, which might contain fine dynamical information of imaged targets. Here, we develop a spatiotemporal scanning technique and realize the measurement of spatiotemporal scanning of TPF ellipsoid with a semiconducting CsPbBr<sub>3</sub> nanosheet. Results have shown that axial size of TPF ellipsoid present linear growth as a function of excitation fluence by using spatial scanning. Furthermore, we have observed that axial size of TPF ellipsoid exhibits inhomogeneous linear growth with time delay by introducing spatiotemporal scanning technique. We attribute this phenomenon to the fact that surface and bulk region of CsPbBr<sub>3</sub> nanosheet have inhomogeneous timescale on TPF decay lifetime. Our results not only provide new insights for spatiotemporal resolving of TPF ellipsoid, but also helpful to promote the development of fluorescence lifetime microscopy technology.

## 1、 Inhomogeneous Trap-State-Mediated Ultrafast Photocarrier Dynamics in CsPbBr<sub>3</sub> Microplates

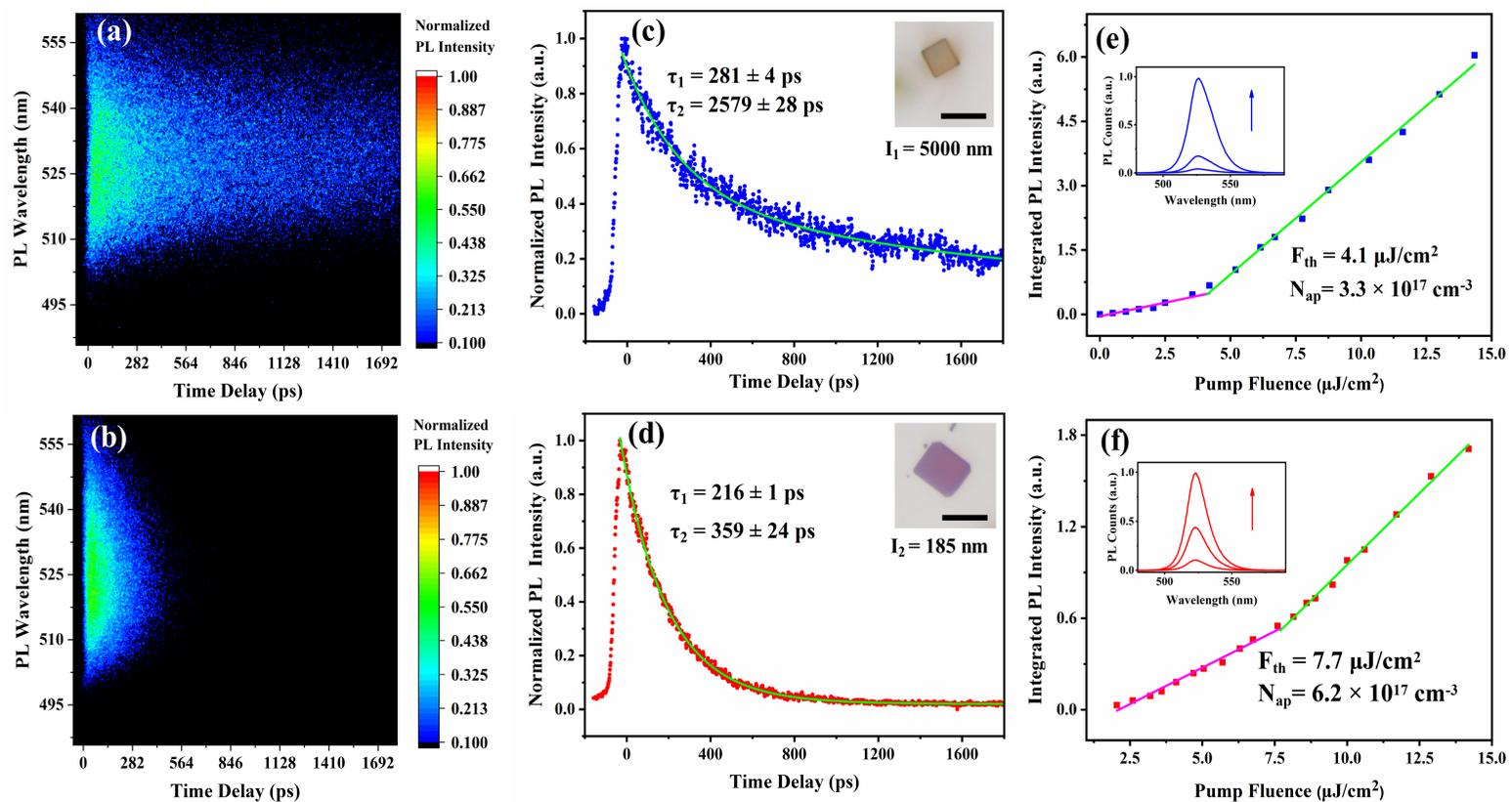
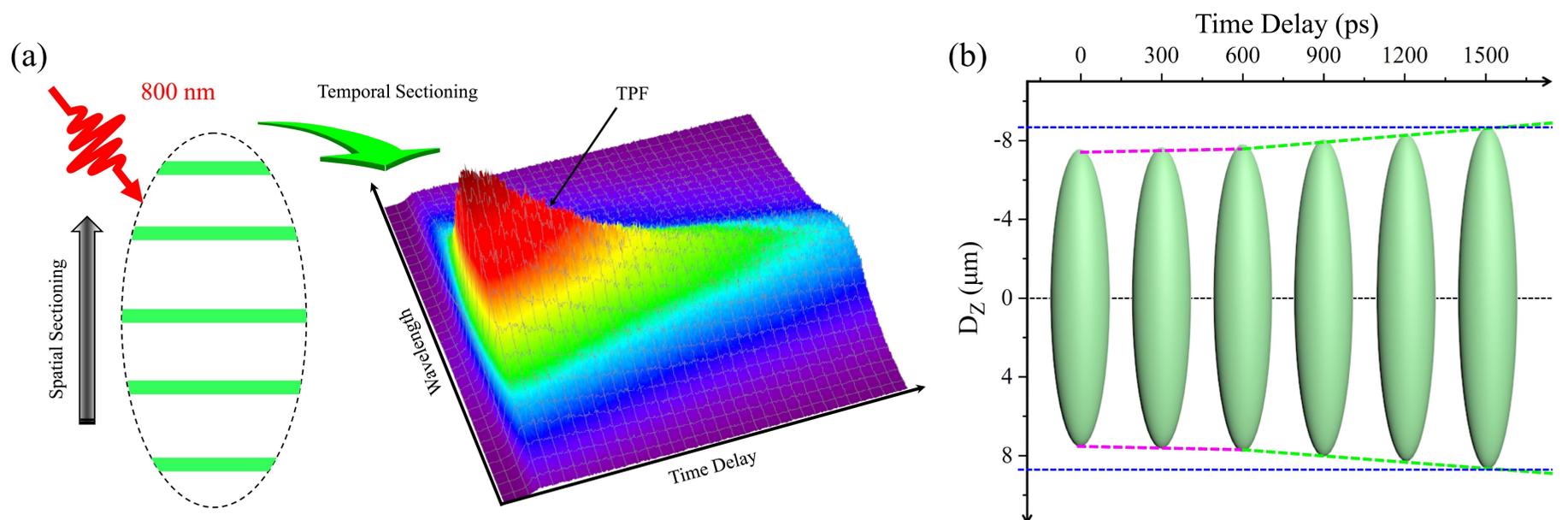


Figure 1. Time-resolved PL spectra (normalized) in (a) a 5000 nm-thick and (b) a 185 nm-thick CsPbBr<sub>3</sub> microplates. (c) Temporal decay (blue squares) of PL intensity at 525 nm extracted from (a); bi-exponential fitting (green solid line) leads to a fast lifetime ( $\tau_1 = 281$  ps) and a slow lifetime ( $\tau_2 = 2579$  ps). (d) Temporal decay (blue squares) of PL intensity at 525 nm extracted from (b); bi-exponential fitting (green solid line) leads to a fast lifetime ( $\tau_1 = 216$  ps) and a slow lifetime ( $\tau_2 = 359$  ps). Corresponding insets show optical microscopy images of two samples with a scale bar of 10  $\mu\text{m}$ .

**Our research indicates that the fast and slow PL lifetime originate from surface region and bulk region because of trap density inhomogeneous distribution.**

## 2、 A New Spatiotemporal Scanning Technique for Two-Photon Fluorescence



We visualize spatiotemporal evolution of TPF ellipsoids along axial direction. At each spatial scanning position along axial direction, time-resolved TPF spectra of an ultrathin luminescent medium is recorded. Then, the TPF ellipsoids are spatiotemporally reconstructed.

We successfully realized the spatiotemporal reconstruction of TPF ellipsoid by a CsPbBr<sub>3</sub> nanosheet. The axial size ( $D_z$ ) of TPF ellipsoid is the smallest at 0 ps. With the TPF time delay extending,  $D_z$  presents a nonlinear growth trend. We attribute this spatiotemporal evolution of TPF ellipsoid to the fact that TPF lifetimes of surface and bulk regions are inhomogeneous.

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### Reference:

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生物电子学国家重点实验室  
STATE KEY LABORATORY OF BIOELECTRONICS

