Second-Harmonic Generation (SHG) Imaging Microscopy with ps Pulsed Lasers

Volker Buschmann, Christian Oelsner, Bita Rezania, Maria Loidolt-Krüger, Rainer Erdmann

PicoQuant GmbH, Rudower Chaussee 29, 12489 Berlin, Germany, www.picoquant.com

PicoQuant

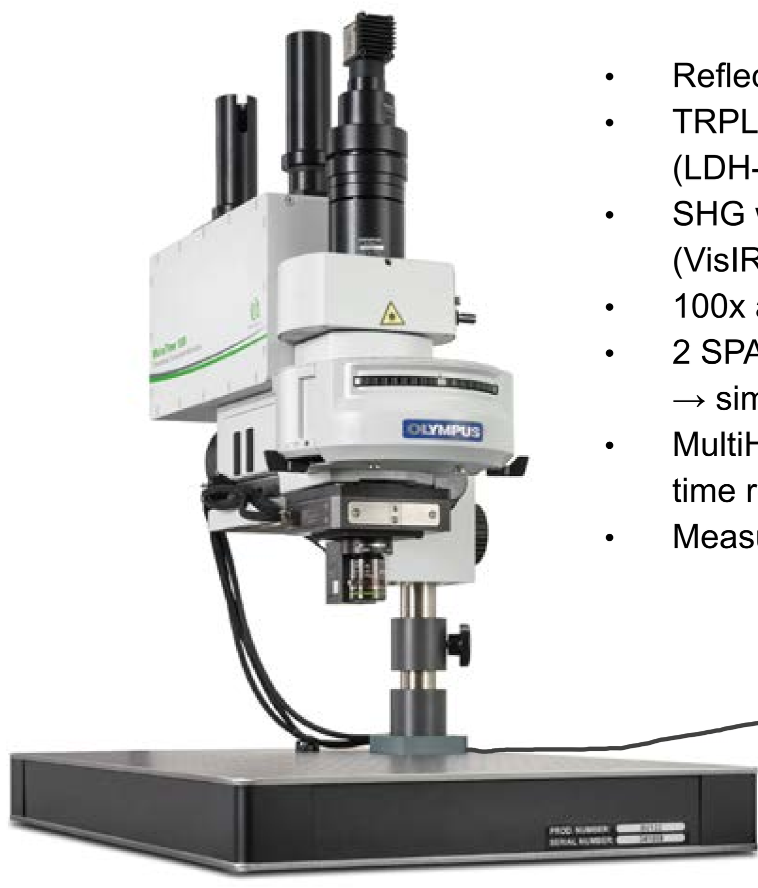
Introduction

Second harmonic generation (SHG) imaging microscopy is a nonlinear optical imaging technique that uses SHG as a contrast mechanism to produce high-resolution images. SHG occurs in materials with non-centrosymmetric crystal structures.
Therefore, SHG imaging has been applied for characterization of 2D semiconductors, transition-metal dichalcogenides (TMDs) such as WS_2 and $MoSe_2$, lithium niobate crystals, PZT thin films, graphene, lanthanides, and even biological tissues. It provides information on the crystal lattice, assesses crystal quality and maps grain boundaries, defects, and mechanical strain. Furthermore, SHG imaging reveals the number of stacked layers as well as their orientation with respect to each other.

SHG imaging is commonly performed with a confocal laser scanning microscope using a high-power fs pulsed laser for excitation. In this study, we establish the use of high power ps pulsed lasers instead, with the aims of decreasing laser safety issues as well as reducing microscope complexity and cost. In addition, ps lasers offer high flexibility and tunability in terms of pulse duration, repetition rates, output power and external triggering.

Measurement Setup

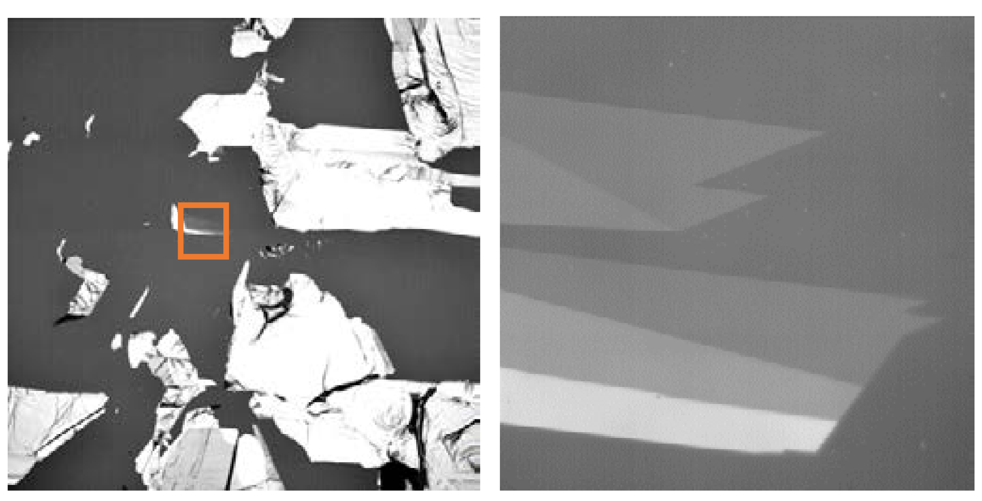
- Reflection image on camera
- TRPL with 640 nm exc @ 20 MHz (LDH-P-C-640B)
- SHG with 1064 nm exc @ 20 or 40 MHz (VisIR 1064) → up to 40 mW
- 100x air IR objective
- 2 SPAD detectors → simultaneous imaging of SHG and TRPL signals
- MultiHarp 150 TCSPC module, time resolution set to 50 ps/ch
- Measurements at ambient condition



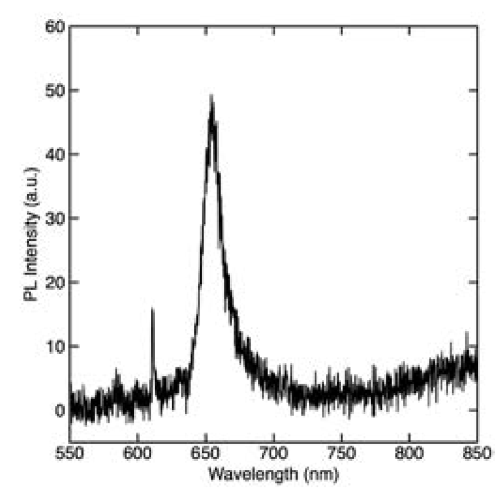
MicroTime 100 BXFM

VisIR 1064

Results: Monolayer MoS_2 on PDMS



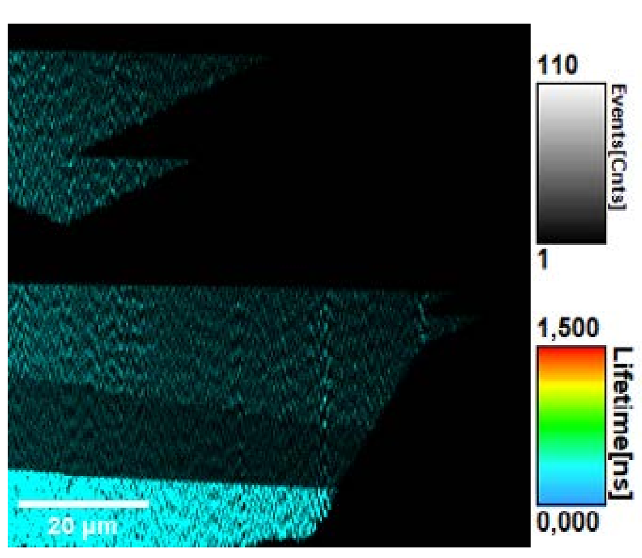
Camera reflection images (4x and 100x objective)



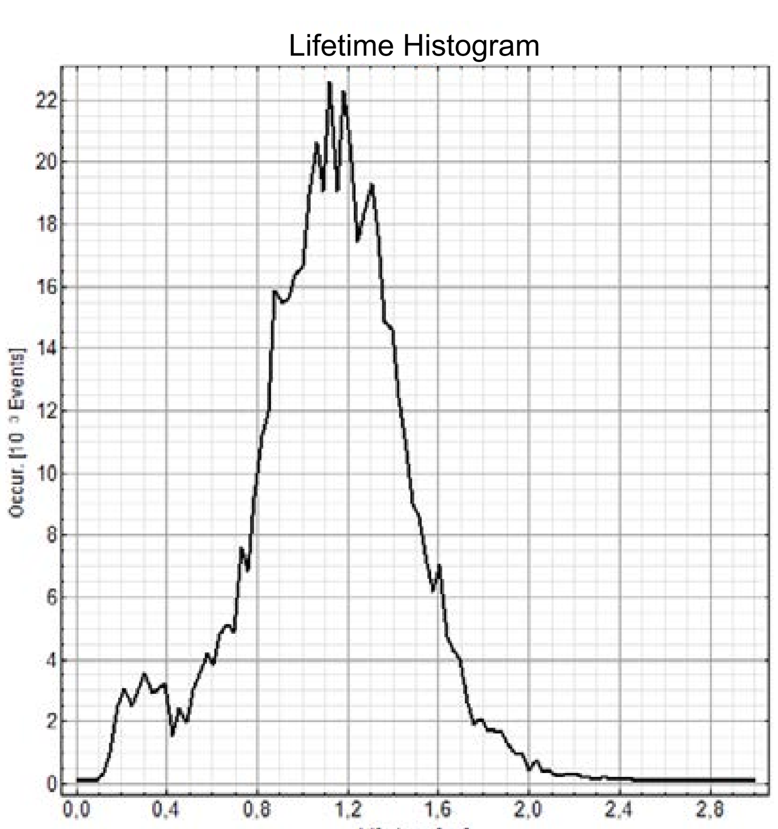
Steady-state emission spectrum

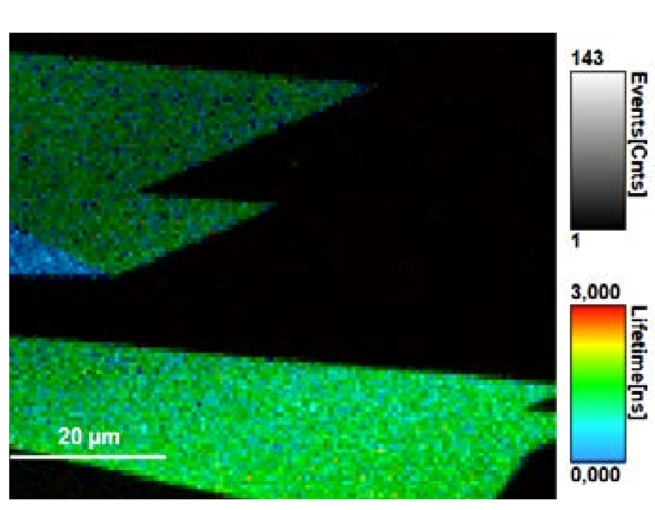
Sample:

- 2D material, monolayer without out-of-plane structure
- In general, the membranes are not flat, but have a 3D-Structure.
- Size of monolayer flakes about 20 µm
- Emission peak MoS_2 ~ 650 nm
- Photoluminescence: BP690/70
- SHG: BP 520/35 + SP 750



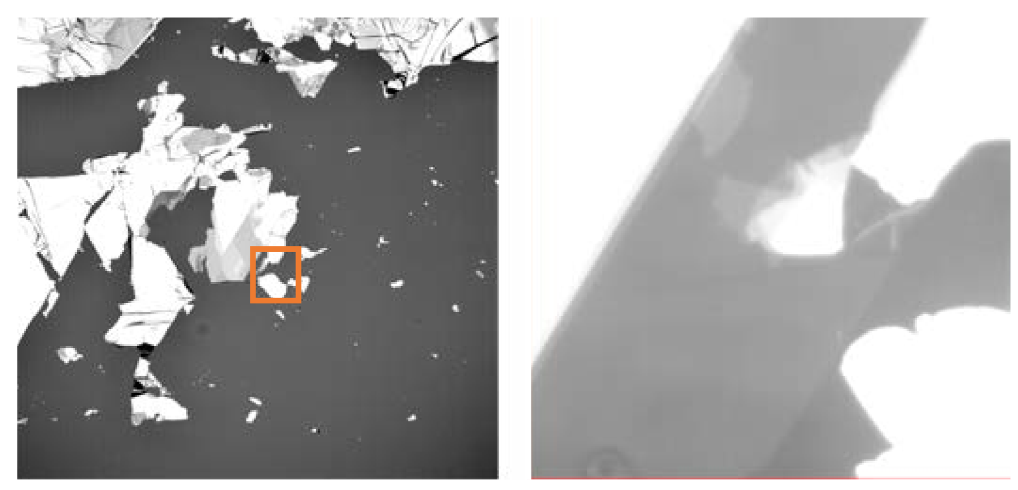
SHG with 1064 nm excitation



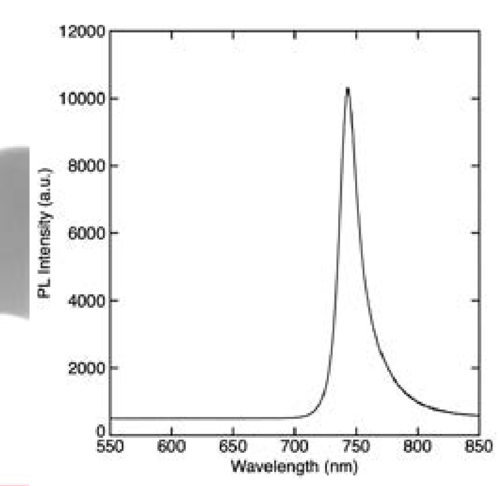


TRPL with 640 nm excitation

Results: Monolayer WSe_2 on PDMS



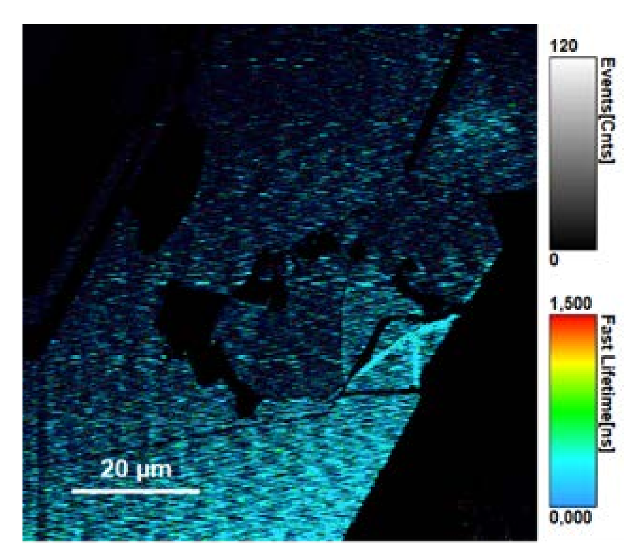
Camera reflection images (4x and 100x objective)



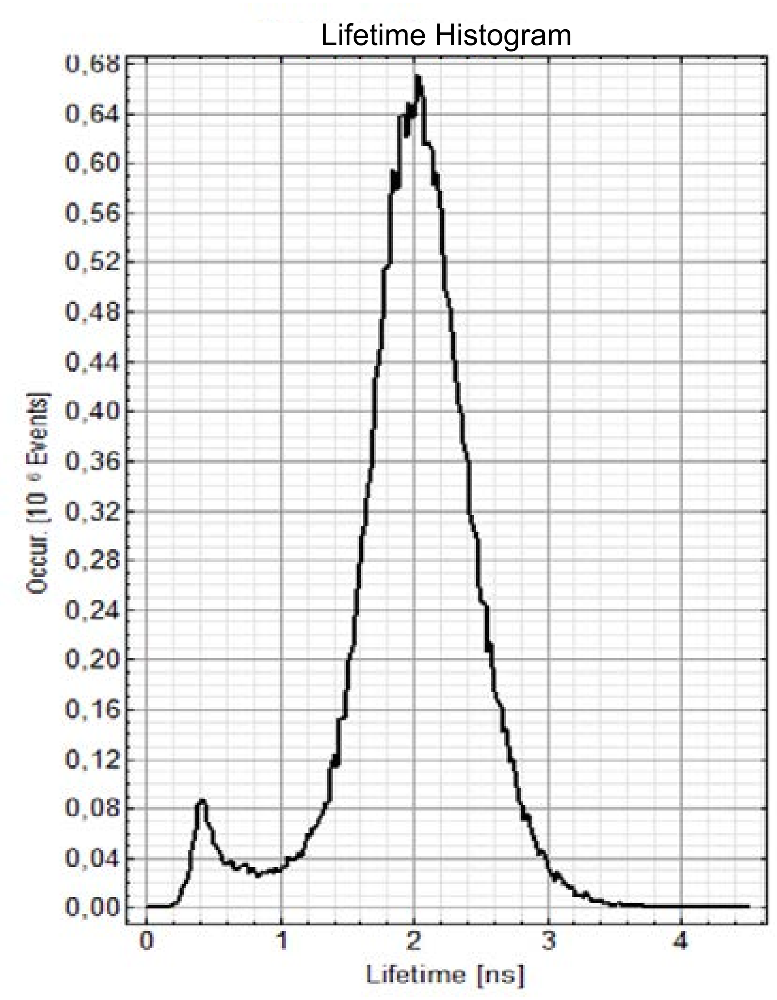
Steady- state emission spectrum

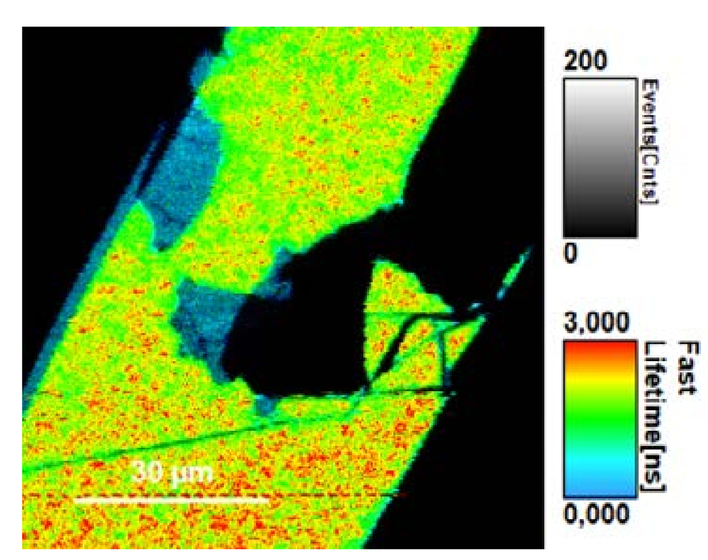
Sample:

- 2D material, flat monolayer without out-of-plane structure
- Size of monolayer flakes about 20 µm
- Emission peak WSe_2 ~ 750 nm
- Photoluminescence: BP775/50
- SHG: BP 520/35 + SP 750



SHG with 1064 nm excitation





TRPL with 640 nm excitation

Conclusion

The results prove that ps pulsed lasers can replace fs lasers for SHG imaging. Moreover, the combination of ps lasers enables the collection of complementary datasets including reflection, SHG and TRPL images collected from the same region of the sample using a single microscope setup.
Thus, the optical properties of the materials can be characterized locally with several techniques to obtain a comprehensive understanding.

Acknowledgement

Samples courtesy of Jin Myung Kim, UC Irvine

References

1) Zheng, H., Wu, B., Li, S. et al. Localization-enhanced moiré exciton in twisted transition metal dichalcogenide heterotrilayer superlattices. Light Sci Appl 12, 117 (2023). https://doi.org/10.1038/s41377-023-01171-w

2) Munkhbat, B., Küçüköz, B., Baranov, D. G., Antosiewicz, T. J., Shegai, T. O., Nanostructured Transition Metal Dichalcogenide Multilayers for Advanced Nanophotonics. Laser Photonics Rev 2022, 17, 2200057. https://doi.org/10.1002/lpor.202200057