

Ultrafast Dynamics of Monolayer MoS₂ Under A Microscope

Using Transient Absorption Microscopy to study excited state kinetics of monolayer molybdenum disulfide (MoS₂)

In this application note, we demonstrate the ability of Ultrafast Systems' TA widefield microscope to measure ultrafast excited state dynamics of MoS₂ monolayer triangles with large area excitation as well as small spot excitation. We demonstrate the microscope's ability to resolve small TA signals (sensitivity) while maintaining diffraction limited spatial resolution.

Introduction to Transient Absorption Microscopy

With the continuing development of nanomaterials, researchers have been getting more interested in studying excited state dynamics in solid state samples. Additionally, it is becoming more important to measure excited state lifetimes at individual sites of the sample instead of a larger macroscopic ensemble. Spatially resolved transient absorption spectrometry (or Transient Absorption Microscopy, TAM) is an important tool for studying such spatially resolved ultrafast dynamics. Typically, these measurements are done by scanning - focusing the monochromatic pump and probe beams into a small spot and repeating the same transient absorption experiment over an array of predefined points on the sample. With this method, even with a high repetition rate laser it normally takes many hours to scan the desired area on the sample.

The Ultrafast Systems Widefield Advantage

The new Ultrafast Systems widefield transient absorption microscope uses a high speed 2D camera to simultaneously image the transient absorption signal from the area of interest with diffraction limited spatial resolution. This approach allows for much faster experiments, typically under an hour even without a high repetition rate laser.

The widefield transient absorption microscope used here is a new product from Ultrafast Systems designed for studying relatively large sample areas (55 μm x 40 μm) with diffraction limited spatial resolution. Our widefield microscope is capable of probing the excited state dynamics over an area of interest in two modes:

1. First, with the excitation being uniformly distributed over the area of interest – area excitation mode;

2. Second, with the excitation being confined to a single, diffraction limited spot on the sample – single spot excitation mode.

In addition to providing spatially resolved transient absorption data, these two approaches are intended to help scientists elucidate sample heterogeneity and spatial distribution of the ultrafast dynamics.

Experimental Methods

The sample used in this application note was CVD-grown monolayer MoS₂ triangles purchased from 6Carbon Technology. The monolayers were deposited on a 2 mm thick sapphire plate to ensure good heat dissipation and light transmission.

The TAM measurements were conducted with a Yb amplified femtosecond laser running at a repetition rate of 5 kHz. The excitation wavelength of 425 nm (pump) was generated with an optical parametric amplifier. A 450 nm long pass filter was used before the detector to block the residual pump light. The 515 nm probe was generated by doubling the frequency of the 1030 nm laser fundamental through a BBO crystal. The pump energy was 35 nJ/pulse in the area excitation mode and 0.05 nJ/pulse in the single spot excitation mode. The probe energy was 27 nJ/pulse. The time window of investigation was 100 ps. The image at each time point was an average of ten measurements with a 0.5 s integration time. The complete experiment duration was 54 minutes. The measurements were conducted under the area excitation mode as well as the single-spot excitation mode. In the area excitation mode, both the pump and the probe were overlapped in a <100 μm spot on the sample. In the single spot excitation mode, the probe remained the same and the pump was focused into a near diffraction-limited spot through an objective.

Results and Discussion

Figure 1 shows the TAM signal obtained using the area excitation mode. The three pieces of MoS₂ triangles were measured simultaneously. The imaged area was 55 μm × 40 μm. Figure 1a, b, and c show the TA image of the MoS₂ triangles at 2ps, 5ps and 20ps, respectively. Figure 1d shows the kinetics extracted from the green, blue and red spots on the three triangles.

It can be seen in the figures that the whole area was rather uniformly excited and each of the spots produced similar kinetic profiles. Around time zero, since the overall spot size is still small and the pump energy is high, the coherent artifact is dominating the measured signal.

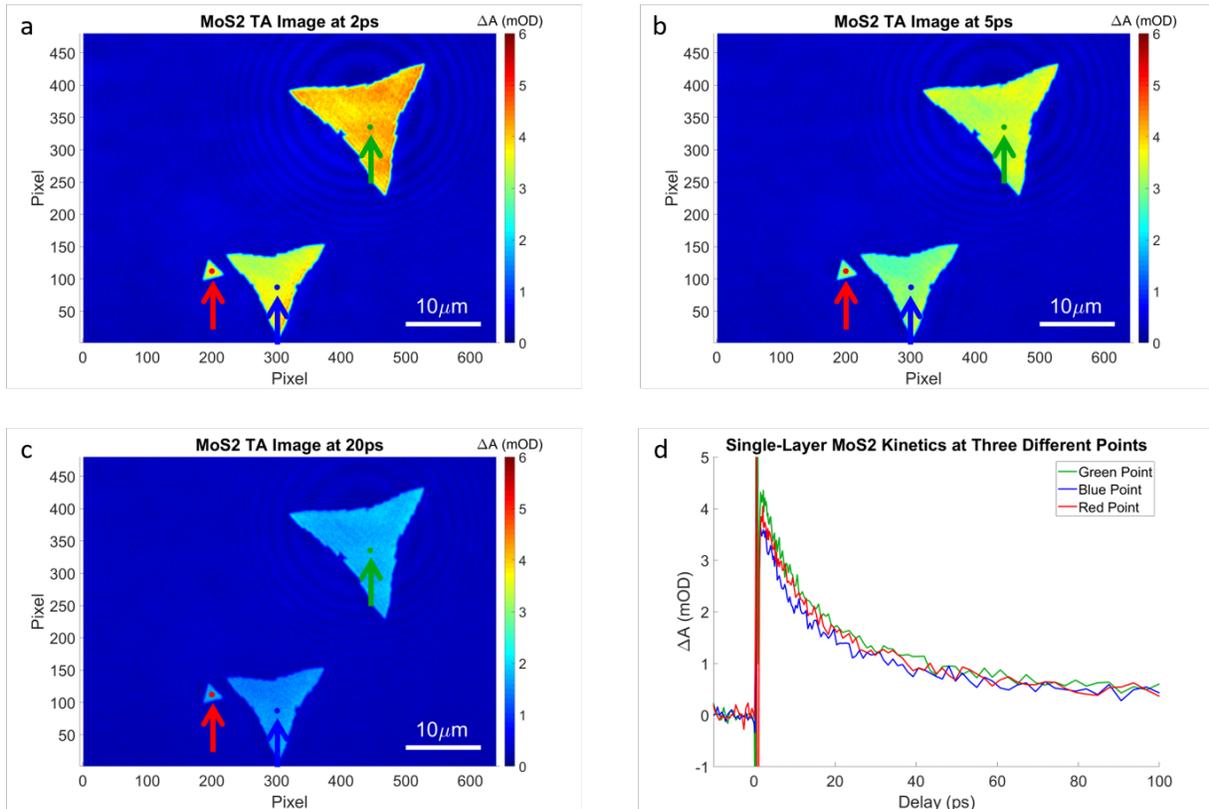


Figure 1 The area-excitation TA measurements. TA image at (a) 2ps, (b) 5ps and (c) 20ps. d) The kinetics at different spots of the three monolayer flakes.

Figure 2 shows the TAM signal obtained using the single-spot excitation mode. A single point on the MoS₂ flake was excited and the whole 2.7 μm diameter spot was probed simultaneously. Figure 2a, b and c show the spatially resolved TA signal at 1 ps, 5 ps and 20 ps, respectively. Figure 2d shows the TA kinetics extracted from the green, blue and red spots on the image.

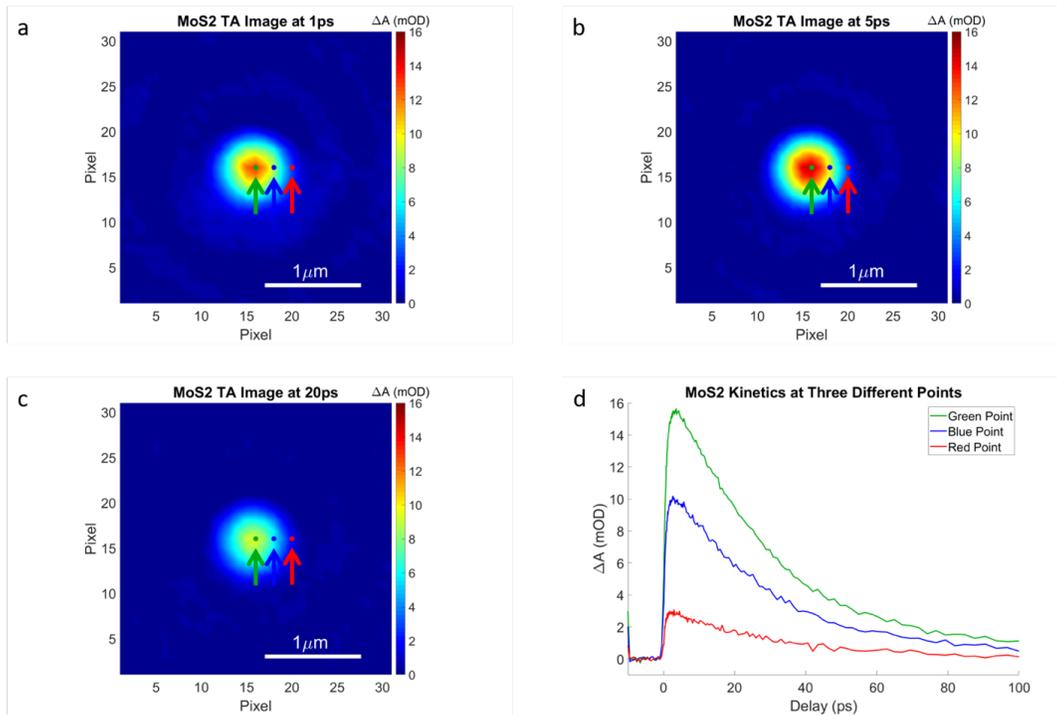


Figure 2 Single-spot-excitation measurement results. TA image at (a) 1ps, (b) 5ps, and (c) 20ps. (d) Comparing the dynamics at three different spots of different spots on the signal image shown in (a)-(c).

The TA signal cross-section extracted from the data are shown in Figure 3. Figure 3a shows the measured TA signal cross-section and Figure 3b shows the normalized results. The spatial FWHM of the signal was ~ 580 nm and did not change over the entire measurement window.

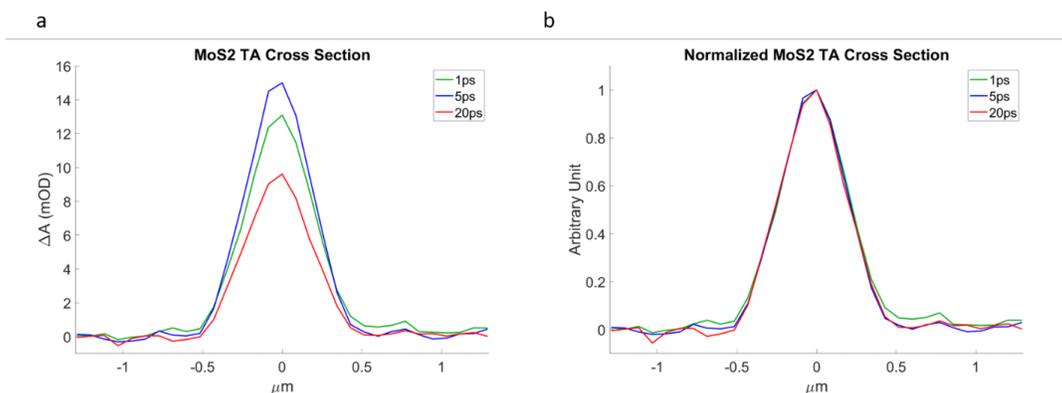


Figure 3 Cross-sections of the TA signal in the single spot excitation mode. (a) Measured raw signal, and (b) Normalized cross-sections at different pump-probe delays.

Conclusion

In this application note, we demonstrated the first commercial transient absorption microscope capable of imaging ultrafast transient absorption signals simultaneously over an area of 55 $\mu\text{m} \times 40 \mu\text{m}$ with near diffraction-limited spatial resolution. Two different modes of excitation were used to study microscopic samples of MoS₂ monolayer flakes. The transient absorption microscope performed well with a low-kHz femtosecond laser and produced good S/N ratios even with small signals. The demonstrated spatial resolution of the microscope is sufficient for studying carrier transport in samples.



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