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## Photoresponse of WSe<sub>2</sub>/MoSe<sub>2</sub> in-plane heterostructure probed by a laser-combined multiprobe spectroscopy

In recent years, transition metal dichalcogenides (TMDC) family such as MoS<sub>2</sub> and WSe<sub>2</sub> has attracted much attention due to their remarkable optoelectronic properties. These monolayers have direct bandgaps in a visible range and ultrafast photo response was recently reported in MoS<sub>2</sub> monolayer [1]. Studying band arrangement, optical/electrical characteristics, and transient carrier dynamics is absolutely necessary. For such purposes, scanning tunneling microscopy (STM) has been used to evaluate the band structure, and time-resolved photocurrent measurement has been used to observe ultrafast carrier dynamics with a prototype of multi-terminal device structure prepared by electrode fabrication techniques such as lithography. However, the wiring process requires physical and chemical treatments of sample. In the case of small (~ $\mu\text{m}$ ) device structures such as a monolayer TMDC island formed on a insulative material, the probe access is rather difficult because of its limited observable area. In addition, each measurement should be done in an identical clean environment because such kind of thin material is sensitive to the adhesives and impurities. An effective approach is to apply multiprobe (MP) STM techniques, in which multiple tips are used to measure, for example, transport characteristics between two desired points, repeatably. However, the simultaneous use of MP techniques with laser technology has not been realized because of the complexities and the handling difficulties.

Here, we present a microscopy method that we have developed by combining MP techniques with optical methods such as Light Modulated scanning tunneling spectroscopy (LM-STs) [2] and Optical Pump-Probe (OPP) STM [3]. To achieve the experiment, we integrated optical zoom lens (VH-Z100T, WD=24mm, Keyence Co., Ltd.) above the MP-STM system, which is used for monitoring probe/sample arrangement as well as for laser positioning and focusing with ~ $\mu\text{m}$  precision[4].

In this experiment, we observed the field-effect transistor (FET) characteristics and photocarriers response in a monolayer island of a WSe<sub>2</sub>/MoSe<sub>2</sub> in-plane heterostructure grown on a SiO<sub>2</sub>/Si substrate (Fig.1a). The FET characteristics and photocurrent were measured in the experimental setup shown in Fig. 1b. Two contact atomic force (c-AFM) tips coated with PtIr<sub>5</sub> were placed in contact with the sample in the MoSe<sub>2</sub> or WSe<sub>2</sub> area to act as the source and drain. As shown in Fig.1c, the spatial distribution of photocurrent ( $I_{ph}$ ) vs drain-source voltage ( $V_{ds}$ ) was measured across the MoSe<sub>2</sub>/WSe<sub>2</sub> interface by changing the focusing spot position (continuous-wave light of 532 nm, ~16  $\mu\text{W}$ ). As a result, we were able to visualize the band structure including the schottkey barrier of probe-sample interface and band shift on MoSe<sub>2</sub>/WSe<sub>2</sub> interface as shown in Fig.1d.

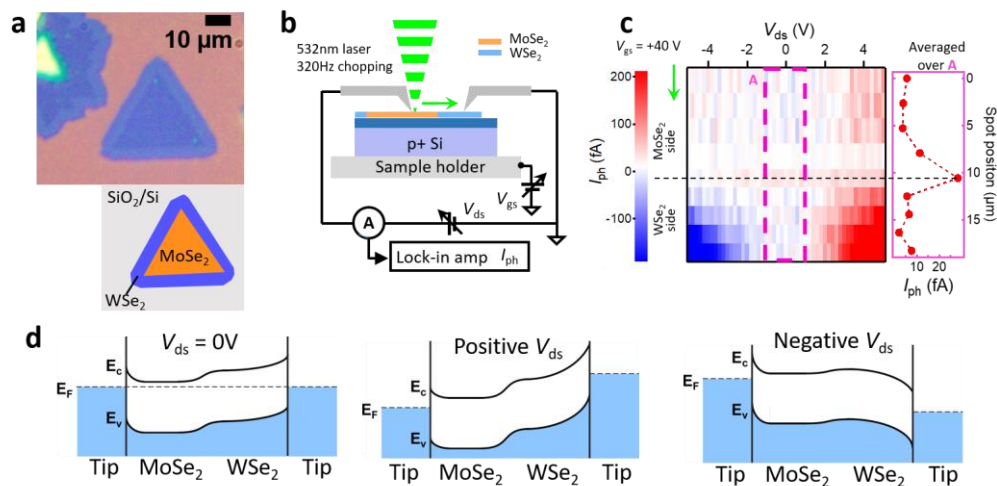
Next, we show the positional dependence of time-resolved measurement realized by the OPP-MP spectroscopy we have developed. The dynamics of photocarriers we have observed in the experiments by using femtosecond pulsed laser with a pulse width of ~150 fs. The probes were arranged across the MoSe<sub>2</sub>/WSe<sub>2</sub> interface, and the laser spot position was set on each probe apex to measure the MoSe<sub>2</sub> or WSe<sub>2</sub> region independently. In the WSe<sub>2</sub> region, four wide range lifetimes were obtained by fitting the signals with exponential functions ( $\tau_1 = \sim 24$  ps,  $\tau_2 = \sim 200$  ps,  $\tau_3 = \sim 20$  ns, and  $\tau_4 = 500$  ns <). On the other hand, in the MoSe<sub>2</sub> region, we obtained two nanosecond components of lifetimes. This difference may come from the effect of the deep-level trap states in the case of MoSe<sub>2</sub>. Details will be discussed in the presentation.

By combining scanning MP microscopy with optical methods such as the Light modulated spectroscopy and OPP methods, we have succeeded in developing a microscopy method useful for measuring electronic structures and photoinduced carrier dynamics in microscopic structures. This technique is expected to play an important role in the researches to develop advanced functional devices with microscopic structures.

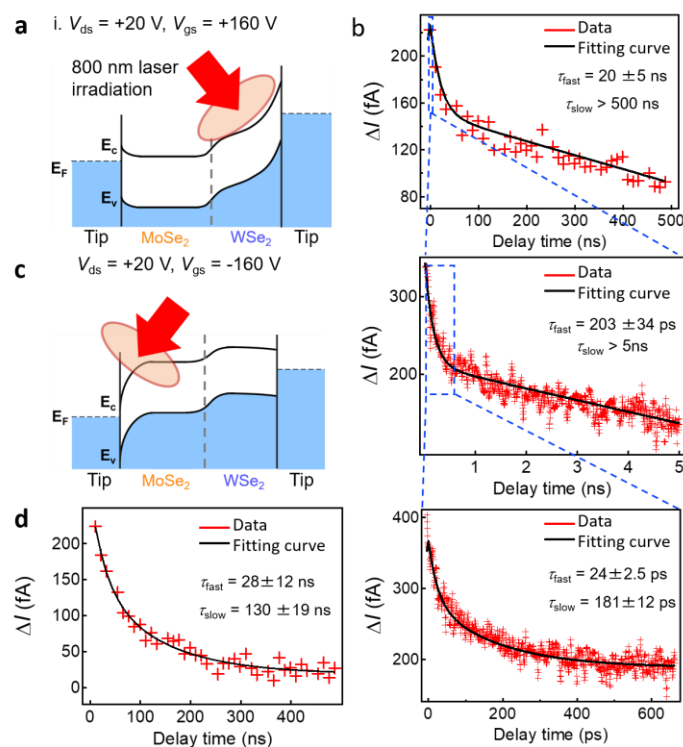
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## Figures



**Figure 1:** Monolayer MoSe<sub>2</sub>/WSe<sub>2</sub> heterostructure sample, **b** Multiprobe photocurrent measurement schematic, **c** Obtained mapping of  $I_{ph}$ - $V_{ds}$  characteristics, and **d** estimated band structures.



**Figure 2:** The carrier dynamics measurement by Optical pump-probe multiprobe technique. Schematics and the results in the cases of irradiating WSe<sub>2</sub> region (**a,b**), and MoSe<sub>2</sub> region (**c, d**)