Photon correlation of photoluminescence emission of a monolayer WS_2

J. Huang[1,*,+], I. Sarpkaya[2,*], J. Lim[1], S-J. Lee[3], X. Duan[4], C. W. Wong[1,+], and H. Htoon[2,+]
[1]*Mesoscopic Optics and Quantum Electronics Laboratory, University of California Los Angeles, CA, USA*
[2]*Center for Integrated Nanotechnologies, Los Alamos National Laboratory, NM, USA*
[3]*Department of Materials Science and Engineering, University of California Los Angeles, CA, USA*
[4]*Department of Chemistry, University of California Los Angeles, CA, USA*
* *equal contribution* [+]*jiahuihuang@ucla.edu, cheewei.wong@ucla.edu, htoon@lanl.gov*

Abstract: We observed the photon bunching of photoluminescence emission from a Tungsten disulfide monolayer by photon correlation measurements. The mixture of spectral diffusion and thermal light emission is considered to explain the bunching signature.
OCIS codes: (250.5230) Photoluminescence; (160.6000) Semiconductor materials; (300.6250) Spectroscopy, condensed matter

1. Introduction

The photoluminescence (PL) emission spectrum of monolayer transition metal dichalcogenide (TMD) materials may contain various spectral broadening effects that affect the spectral purity of the emission. One of the external inhomogeneous broadening effects is the spectral diffusion (SD) caused by the Stark shift of energy level due to the fluctuation of environmental spin and charge around the emitter leading to fast back-and-forth wandering of PL spectrum [1]. The microscopic origin of spectral diffusion in the emission of 2D materials must be understood and suppressed for efficient use of them in optoelectronic and quantum photonics applications. The SD measurements could provide information about the local environment of light emitters. A novel technique based on correlations of photons within a spectral window narrower than the SD broadened PL spectrum has been demonstrated to extend the SD studies to the nanosecond time scale for the case of individual semiconducting quantum dots [2]. This technique was also applied to individual SWCNTs and nanosecond spectral diffusion time measured in the PL emission of carbon nanotubes [1,3]. In this paper, we applied the photon correlation spectroscopy for the first time to one of the emerging 2D materials (monolayer WS_2) to reveal its SD dynamics.

2. Experiment

The second order correlation measurements were performed with a Hanbury Brown and Twiss Interferometer setup under 532-nm cw laser excitation (Fig. 1a). We prepared a monolayer WS_2 sample grown by the chemical vapor deposition (CVD) on a sapphire substrate and performed PL mapping to probe the uniformity of the flake in the sample. We focus the laser beam onto the brightest spot based on the PL mapping (Fig. 1b). The center of the PL spectrum is approximately at 630-nm at room temperature. A 545-nm long pass filter and a bandpass filter centered at 625-nm with a 50-nm full width half maximum (FWHM) were used to eliminate any nonrelevant laser or scattering lights (Fig. 1c). The filtered PL spectrum is then sent to a photon correlation setup to measure the $g^2(\tau)$. A 628-nm tunable bandpass filter (14-nm FWHM) was used to extract only left-hand side (LHS) of the whole PL spectrum. The $g^2(\tau)$ of the LHS spectrum is then measured at different locations on the flake (bright and dark spots on the PL map). We repeat the same measurement at the cryogenic temperature (5K).

$g^2(\tau)$ measurements for different spectral windows are also performed to probe the effect of the spectral window on the SD. $g^2(\tau)$ is measured for entire PL spectrum (filtered with 50-nm bandpass filter) and LHS spectrum with 15, 7 and 3 nm widths from the same position on the flake.

3. Results and discussion

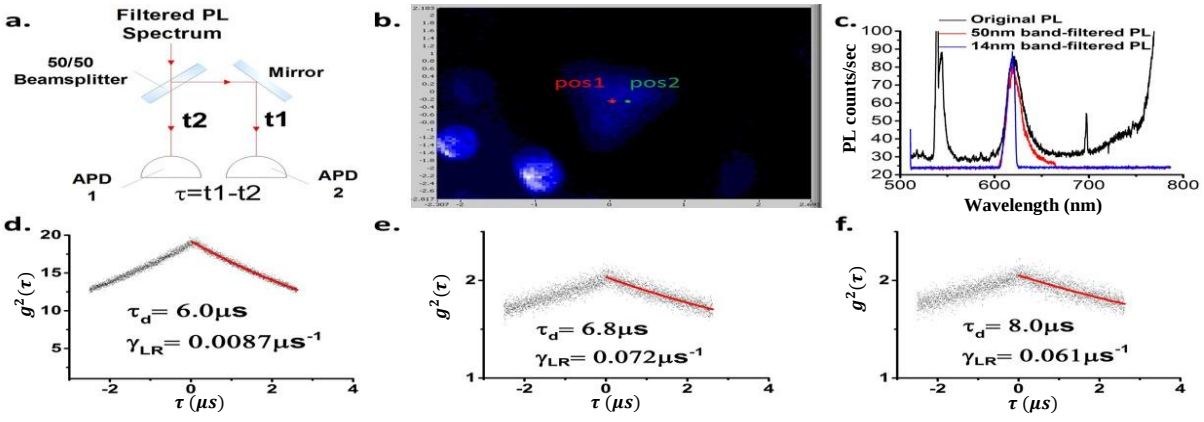

Fig. 1 (a) Schematic of Hanbury Brown and Twiss (HBT) setup for second order photon correlation measurements. (b) Confocal PL mapping of a monolayer WS_2 flake. (c) PL spectra of a monolayer WS_2. (d) $g^2(\tau)$ for spectrum in b. at position x=0.218 y=-0.251 (Bright spot, position 1) on the flake. (e) $g^2(\tau)$ for spectrum in c. at position x=0.218 y=-0.251 on the flake. (f). $g^2(\tau)$ for spectrum in c. at position x=0.486, y=-0.251 (Dark spot, position 2). The red solid lines are fitting curve with $g^2(\tau) = 1 + \left(\frac{\gamma_d}{\gamma_{LR}} - 1\right)\exp(-\gamma_d\tau)$, where γ_{LR} is the crossover rate depending on the spectral window size and $\gamma_d = \frac{1}{\tau_d}$ is the SD rate. Laser power on the flake is 2 μW. All data are recorded at 5K.

$g^2(\tau)$ is measured at different locations on a monolayer WS_2 sample flake at 5K. The PL mapping in Fig. 1b shows that the flake is brighter at the center (x=0.218, y=-0.251, position 1) and is darker away from the center (x=0.486, y=-0.251, position 2). $g^2(\tau)$ of 50-nm and 14-nm spectrum at bright location are showed in Fig. 1d and Fig. 1e respectively. Corresponding $g^2(\tau)$ of 14-nm spectrum at dark location is showed in Fig. 1f. The results show that for the bright emitter, the wider the spectral window is, the smaller τ_d and γ_{LR} are, which means that the bunching peak decays faster and its height becomes larger. The bright emitter also shows smaller τ_d compared to dark emitter for the case of 14 nm filtered spectrum.

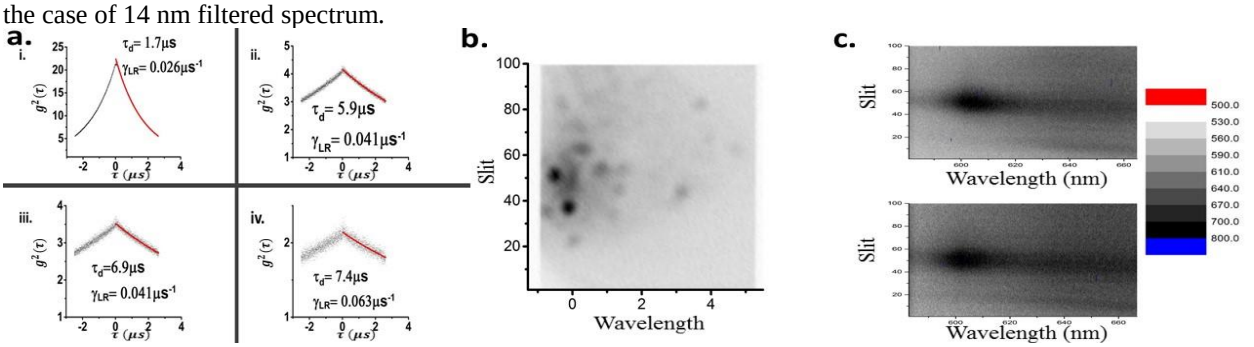

Fig. 2 Spectral window size dependence of the second-order correlation function for the same position on the same flake. Corresponding $g^2(\tau)$ histograms for entire PL spectrum (a-i), 14-nm filtered LHS spectrum (a-ii), 7-nm filtered LHS spectrum (a-iii) and 3-nm filtered LHS spectrum (a-iv). All cases show the photon bunching and SD time increases with the decreased spectral window-size. All data are taken at room temperature. Laser power on the sample is 2 μW. The red solid lines are the fitting curve with $g^2(\tau) = 1 + \left(\frac{\gamma_d}{\gamma_{LR}} - 1\right)\exp(-\gamma_d\tau)$. (b)Wide field image of one of the WS2 flakes at 5K. "Wavelength" and "slit" in horizontal and vertical axis mean position. (c) Spectral image of two bright spots in (b). "Slit" in vertical axis mean position.

$g^2(\tau)$ is also measured for different spectral window sizes at a single position on the flake. Pronounced photon bunching signature is observed with the wider PL spectrum (Fig. 2a-i). τ_d and γ_{LR} values increase for narrower filtered spectrum (Fig. 2a-ii, iii and iv) and show the same trend observed in Fig. 1d-e. Since τ_d has to be independent of the spectral window-size and $\gamma_{LR} \to 0$ for very narrow spectral window size so that the narrow spectral window will generate a pronounced SD bunching signature [2], which is opposite to our results. Wide field spectroscopy shows some spatially localized spots on one of the flakes (Fig. 2b) but such spots do not have spectrally localized sharp peaks (Fig 2c). We also observed a very similar μs bunching behavior when we sent a white light (thermal light) beam with intensity similar to the PL of WS_2 to the correlation setup. Based on the above observation we conclude that the observed photon bunching behavior of our WS_2 sample is due to the thermal light nature of the monolayer WS_2 emission. Thermal light emission creates a pronounced photon bunching signature, which is superimposed with the SD bunching signal implying that SD is masked by the thermal bunching that has a longer decay time constant (μs timescale). The bunching signature of thermal light originates from its large intensity fluctuation. The brighter spots on the flake and wider spectral window would have more fluctuations. So, by treating the WS_2 PL as a thermal light could explain the bunching trend in Fig. 1, 2. To extract the SD characteristic from the thermal light bunching profile, a cross-correlation measurement needs to be applied [2]. For an auto-correlation measurement, both thermal light and SD shows the bunching signature so that SD is masked by the thermal light bunching profile. But for a cross-correlation measurement that photons from two separated spectral windows are correlated, SD could lead to antibunching signature while the thermal light still shows bunching signature. Two features can be separated, and the SD characteristic time can be extracted from the antibunching near $\tau = 0$ in the overall thermal bunching profile. We are currently performing this experiment.

In conclusion, we show the SD characteristic of monolayer WS_2 is masked by the pronounced bunching due to its thermal light emission behavior, which is different from carbon nanotube or quantum dot samples whose PL emission are coherent or single photon antibunched. Therefore, to extract SD from the thermal light bunched emission of the monolayer WS_2, the cross-correlation measurement needs to be applied.

4. References

[1] W. Walden-Newman, *et al*., Quantum light signatures and nanosecond spectral diffusion from cavity-embedded carbon nanotubes. *Nano Lett*., **12**, 1934-1941 (2012)
[2] G. Sallen, *et al*., Subnanosecond spectral diffusion measurement using photon correlation. *Nat. Photon*. **4**,696-699 (2010)
[3] I. Sarpkaya *et al*., Prolonged spontaneous emission and dephasing of localized excitons in air-bridged carbon nanotubes. *Nat. Commun*. **4**, 2152 (2013)