Supplementary Information for Insight into Exciton Polaritons of Two-Dimensional Transition Metal Dichalcogenides with Time-Resolved Spectroscopy

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1 Time-Resolved Spectroscopy Techniques, Additional Details

1.1 Time-Resolved Photoluminescence Spectroscopy

TRPL methods typically use spectrally-narrow pump lasers to excite the material well above resonance, where the resulting emitted signal is resolved by spectrally filtering out the pump. The temporal resolution of TRPL spectroscopy varies by orders of magnitude depending on the detection methods used with trade-offs between temporal resolution, sensitivity, and temporal sweep range. Time-correlated single photon counting (TCSPC) [1] measures emitted photons using a single photon detector that sends an electrical signal to a timing circuit for each detection event. The timing circuit consist of either separate time-to-amplitude and analog-to-digital conversion steps or direct time-to-digital conversion to time the arrival of these electrical signals, which are used to build up a histogram of the arrival time of emitted photons as referenced to the laser pulse arrival time. This technique is relatively easy to employ and highly sensitive to low light levels, where the excitation power density is low enough that the emission probability per pulse is much less than one. The fast measurement acquisition speed has led to its frequent use in fluorescence lifetime imaging. However, the $10 - 100s$ *ps* resolution of typical single photon avalanche photodiodes (SPADs) and photomultiplier tubes limits the temporal resolution and short-wave IR (SWIR) SPADs have relatively low quantum efficiency. In contrast, streak cameras [2] employ a streak tube, which uses a time-varying field to accelerate electrons from a photocathode towards a phosphor screen, to create a streak image on a CCD array where one dimension

represents time and the other often represents wavelength. While this technique can achieve $\sim$ ps resolution, Streak cameras are typically expensive, require significant maintenance to ensure reproducibility, and there are also trade-offs between temporal resolution, sensitivity, and temporal sweep range. Sub-ps resolution TRPL requires detection techniques that employ nonlinear optical (NLO) sampling, like optical Kerr effect [3], or frequency mixing, including up-conversion [4]. While up-conversion TRPL offers higher signal-to-background compared to OKG, it is frequently used in conjunction with a monochromator and PMT, which reduces this technique to a single emission wavelength measurement and thereby increases the measurement time for acquisition of time-resolved spectra. Additionally, the phase matching requirements often requires the NLO medium to be angle tuned to change the detection wavelength. However, for a SWIR mixing pulse, broadband up-conversion TRPL can be realized in type-II KDP or BBO and detected via a spectrograph and CCD [5].

1.2 Pump Probe Spectroscopy

Time-resolved Pump Probe spectroscopy is a nonlinear spectroscopy technique that uses a sequence of two laser pulses (Fig. 3b) to measure the third-order material response of a system as a function of wavelength and time delay ($S_{PP}(\lambda, T)$) [6]. The first laser pulse, the so-called 'pump' pulse, is spectrally narrow band in order to selectively-excite a specific electronic transition. This initial pump pulse yields two light-matter interactions ($S_{PP} \propto |E_1|^2$), either preparing excited populations, similarly to in TRPL, or additionally preparing coherent superposition states. In contrast to TRPL, PP employs a second pulse (E_2) delayed from the pump pulse to monitor the evolution of the excited states as a function of a mechanically-scanned time delay. The resulting PP signal is linearly proportional to the weak second pulse ($S_{PP} \propto E_2$), where E_2 effectively 'probes' the evolution of the populations prepared by the pump pulse as a function of a scanned time-delay, T.

PP spectroscopies typically rely on pulses supplied by Ti:Sapphire regenerative amplifiers, which produce intense pulses centered around $\lambda_o = 800\ nm$ with pulse durations ranging from $\sim 40 - 100\ fs$ and $\sim 1s\ kHz$ repetition rates, though several studies discussed in this prospective have instead used fs oscillators with $\sim 76\ MHz$ repetition rates. These high-intensity pulses are necessary to generate frequency-tuned pump pulses, generated using optical parametric amplifiers (OPA), as well as the broadband probe, generated through supercontinuum white light generation (WLG) [7]. A typical PP apparatus splits the fundamental laser beam into pump and probe paths before both beams are overlapped at the sample by focusing optics; the pump is generated in an OPA and is modulated by a mechanical chopper, whereas the probe is produced in a white light generating crystal and is delayed from the pump pulse via a mechanical delay stage. In PP microscopy (Fig. 3e) the pump and probe typically propagate collinearly into the microscope objective where the measured signal can be collected in transmission or in reflection; in either case, pump pulse scatter is removed either through polarization selection, or through spectral or spatial filtering. In PP experiments using long focal length focusing elements (Fig. 3e), the pump and pulse beams can propagate collinearly or noncollinearly, which has the advantage of requiring less invasive filtering of the pump pulse. The PP

signal is finally extracted by measuring the probe in a spectrometer with the pump 'on' and 'off' and calculating the differential signal ($(I_{2,on} - I_{2,off})/I_{2_{o}ff}$) as a function of time delay, T, yielding a wavelength- and time-dependence differential signal ($S_{PP}(\lambda, T)$). Transient Reflectance ($\Delta R/R$) and Transient Absorption ($\Delta T/T$) are PP spectroscopies specifying that the signal measured is the reflected or the absorptive component. The excitation frequency is set by the pump pulse spectrum while the detection frequency range is set by the probe pulse and the frequency resolution is determined by the spectrometer diffraction grating and camera pixel density. The temporal resolution of a PP experiment is typically limited by the pump pulse, where the spectral bandwidth of the pump provides a lower limit on the pump's temporal duration when fully compressed ($\Delta\tau_{pump} \approx 10s\ fs$) though many PP experiments do not perform pump compression, yielding typical instrument response function (IRF) times of $\tau_{IRF} \geq 100\ fs$.

1.3 Multidimensional Coherent Spectroscopies

Multidimensional Coherent Spectroscopies (MDCS) are Fourier Transform based nonlinear spectroscopies, including Two-dimensional Electronic Spectroscopy (2DES) and Fluorescence-detected 2DES (F-2DES) (Fig. 3c), which measure the same third-order material response as PP but with simultaneously high frequency and temporal-resolution [8, 9]. This is accomplished with a sequence of three spectrally-broadband laser pulses ($S_{2DES} \propto E_1 E_2^* E_3$), where a second 'pump' pulse (E_2^*) is introduced and scanned along a time delay t_1 with high phase-stability. Fourier transformation with respect to t_1 yields the excitation frequency axis ($\hbar\omega_1$), eliminating the need for spectrally narrow band pump pulses as in PP spectroscopy. In 2DES, the detection axis ($\hbar\omega_3$) is directly measured in a spectrometer, similarly to PP, where the differential signal is typically presented as a function of two frequency axes and t_2 ($S_{2DES}(\hbar\omega_1, t_2, \hbar\omega_3)$). F-2DES introduces a fourth pulse (E_4^*) at time delay t_3 to prepare the system into a population state and detects radiated fluorescence signal (Fig. 3c); in this so-called action-detected method, the fluorescence signal is typically measured using lock-in modulation detection with an avalanche photodiode and the detection frequency axis is resolved via Fourier transformation of t_3 ($S_{F-2DES}(\hbar\omega_1, t_2, \hbar\omega_3)$) [8]. Both 2DES and F-2DES methods allow for simultaneously-high frequency and temporal resolution, where use of transform-limited spectrally-broadband pulses typically yields $\tau_{IRF} \leq 20\ fs$. While 2DES and F-2DES measure the same third-order material response, F-2DES can achieve a higher spatial resolution than 2DES due to the fourth-order dependence on pulse electric field ($S_{F-2DES} \propto E_1 E_2^* E_3 E_4^*$), though differences between measured responses of the two techniques can arise due to nonradiative relaxation following E_4, as the fluorescence decay is not time-resolved [10].

The spectrally-broadband pulses used in MDCS are typically generated from one or multiple OPAs and are each compressed near the time-bandwidth transform limit. Though 2DES can be performed in a number of different beam geometries ranging from all-collinear to a box-CARS fully noncollinear geometry, F-2DES is typically performed with all four pulses propagating collinearly, which are then integrated into a high NA microscope objective [8] with reported spatial resolutions of $d_{F-2DES} \simeq 260\ nm$ [11]. The advantage of high temporal and frequency resolution obtained through MDCS comes with the trade-off of increased experimental acquisition times compared to TRPL and

PP techniques. In the case of F-2DES, which scans three time delays and requires complex phase cycling techniques, experimental acquisition times can approach a day [11]. Despite the increased acquisition times of MDCS, these techniques are well suited to studying the sub-100 fs dynamics of EPs in TMDs, where the high frequency resolution additionally allows for clear distinction of signals in spectrally congested and heterogeneous systems.

Table 1 Time-Resolved Studies of TMD Polaritons. Summary of time-resolved studies discussed in this prospective grouped by material type. Polaritons are formed with the coupling target, which ranges from intralayer neutral excitons (X), trions ($X^{\pm}$), biexcitons ($XX^{o,\pm}$), and polarons, as well as interlayer excitons (ILX). Where provided, we indicate the reported Purcell Enhancement factor (F_P), Rabi Splitting (Ω_R), experimental instrument response function (τ_{IRF}), and experimental Temperature (T).

Ref.	Material			System	F_P	Ω_R (meV)	Exp. Technique	τ_{IRF} (ps)	T (K)
[12]	WS_2	ML	X	FP		80	TRPL	≤ 4	RT
[13]		3L NT	X	CMR			PP^A	< 0.2	
[14]		ML	X	PhC		40.2	PP^R		RT
[15]		$N > 10$	X	LSPR		$250^{\dagger}$	$PP^{R,A}$		RT
[16]		ML	X	FP		24	PP^R		200
[17]		ML	X	PGC		183, $273^{\ddagger}$	TRPL		RT
[18]		ML	X	PSLR			PP^R	~ 0.2	RT
[19]		FL	C	LSPR		293	PP^R		RT
[20]		ML	X	$LSPR^g$		50	PP^R		RT
[21]		ML	X,X^-,XX^-	PSLR		88, 80, 70	PP^R	~ 0.2	4, RT
[22]		$MLS^{N=1-4}$	X	FP		34 ± 2, 52 ± 3, 62 ± 2, 71 ± 3	$PP^{R,k}$		RT
[11]	WSe_2	$MLS^{N=2}$	$X^{(ph)}$	FP			F-2DES	0.020	RT
[23]		$MLS^{N=2}$	X	FP			F-2DES	0.020	RT
[24]		ML	X	SiN NA	≥ 20		TRPL		5
[25]		ML	X, $X-polaron$	FP^{OT}		14, ~ 10.3	PP^A		4.2
[26]	MoS_2/hBN/WS_2	HS	X	FP		~ 26	$PP^{R,k}$		
[27]	$MoSe_2$/WSe_2	HS	ILX*	FP^{OT}	1.8 ± 0.3	$0.195 \pm .009^o$	TRPL		4.2
[28]		HS	ILX*	SiN^g	2.4		TRPL		5
[29]		HS	ILX*	PhCC	~ 65		TRPL	~ 450	5
[30]	MoS_2/WS_2	HS	ILX*	PGC	12-45		TRPL	227	5, RT
[31]	MoS_2	BL	ILX	FP		21.4	Pumped-R_c, PP^R	~ 0.2	3.5

TRPL - Time-resolved photoluminescence; PP - Pump Probe Spectroscopy, ^{R} - Reflectance, ^{A} - Absorption, ^{k} - k-space detected; F-2DES - Fluorescence-detected Two-Dimensional Electronic Spectroscopy; TNFI - Time-resolved Near-field Imaging. ML - Monolayer; 3L NT - three layer nanotube; FL - Few-layer; MLS^N - N layer ML Superlattice; Slab - $N > 100$; HS - Heterostructure; BL - homoBiLayer. FP - Fabry Perot, ^{OT} - Open and Tunable; CMR - intrinsic Cavity Mode Resonance; PhC - Photonic Crystal; LSPR - Localized Surface Plasmon Resonance; SiN NA - SiN Nanoantennas, ^{g} - Grating; PhCC - Photonic Crystal Cavity; PSLR - Plasmonic Surface Lattice Resonance; PGC - Plasmonic Gap Cavity. * - Weak light-matter coupling; † - Peak separation in transient response; ‡ - Determined with dark field scattering, ^{o} - Calculated.

Table 2 Exciton-Polariton Nonlinearities from steady-state and time-resolved measurements. Summary of polariton-polariton nonlinear coefficients ($g_{pol-pol}$). Where reported, the constituent exchange-interaction (g_{ex-ex}) and saturation (g_{sat}) coefficients, or alternatively the multi-particle interaction (β), linewidth broadening (α), and saturation density (n_s) strengths, are given. Reported nonlinear interaction strengths for *uncoupled* MLs are for excitons in uncouple material.

Ref	Exp. Technique	Material			$g_{pol-pol}$ ($\mu eV\mu m^2$)	g_{ex-ex} ($\mu eV\mu m^2$)	g_{sat} ($\mu eV\mu m^2$)	β ($\mu eV\mu m^2$)	α ($\mu eV\mu m^2$)	n_s (μm^{-2})
[32]	Pumped R_c	WS_2	*uncoupled* ML	X	6×10^{-4}	0.06	0.005			
[22]	PP^A		ML	X		0.055 ± 0.015	0.110 ± 0.035			
			MLS$^{N=3}$	X		0.01 ± 0.008	0.04 ± 0.01			
[21]	PP^R		*uncoupled* ML	X					0.07 ± 0.02	$(8 \pm 1) \times 10^5$
				XX^-					0.5 ± 0.1	$(2.0 \pm 0.2) \times 10^4$
			ML	X	$0.08 - 0.14$				0.21 ± 0.05	$(3.0 \pm 0.5) \times 10^5$
				X^-	0.2					
				XX^-	$0.8 - 4$				0.36 ± 0.11	$(5.3 \pm 0.8) \times 10^3$
[33]	Pumped R_c	WSe_2	MLS$^{N=3}$	X	$\sim 10.0 \pm 4.2$					
				A^o_{2s}	$\sim 46.4 \pm 13.9$					
[34]	Pumped R_c	MoS_2	BL	ILX	0.86					
[35]	Pumped R_c	$MoSe_2$	ML	X				0.096	0.094	0.04
[36]	Pumped R_c		ML	X		$2 - 5^2$, 0.01^3				
				X^-			37 ± 3			
[25]	PP^A		*uncoupled* ML	X	0.08^6					
	PP^A		ML	X	0.01					
				X-Polaron	0.5					
[37]	Pumped R_c		*uncoupled* ML	X		1.4	*minor*			
			ML	X	$\sim 0.04 - 1.6$	1.0 ± 0.4, 1.6^6	*minor*			
[38]	Pumped R_c		ML	X	2.2 ± 1.6^4, 4.3^5	2.16 ± 0.5^4, 1.6^5				
[31]	Pumped R_c		BL	X	$\sim 10 \pm 0.2$					
				ILX	$\sim 100 \pm 2$					
[39]	Pumped R_c	$WS_2/MoSe_2$	HS	X	$0.02 - 0.04$					
				Moiré-X	$4 \times g^X_{pol}$					

Pumped R_c - Reflectance Contrast with pumped polariton denisty; PP - Spectroscopy, ^{R} - Reflectance, ^{A} - Absorption. ML - Monolayer; MLS^N - N uncoupled stacked MLs; BL - Bilayer; HS - Heterostructure. X - ground state, charge neutral A exciton (A^o_{1s}); X^- - negatively charged trion; XX^- - negatively charged biexciton; A^o_{2s} - first excited Rydberg state; ILX - Interlayer exciton. ^{2,3} Polariton density $n_{tot} \sim 10^3$ μm^{-2}; $\sim 10^5$ μm^{-2}. ^{4,5} $|X|^2 \sim 0.48$, ~ 0.33. ^{6} Calculated value.

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