

**Fig. 1** Typical time orderings of the excitation pulses to generate different types of time/frequency domain 2D spectra. (a) Zero-quantum spectrum for an inhomogeneously broadened V-shape three-state system with a ground state and two mutually dipole-forbidden excited states. (b) One-quantum spectrum for an inhomogeneously broadened three-state V system. (c) Double-quantum spectrum for a three-state ladder system where a doubly excited state exists.

two-dimensional (2D) limit. Secondly, the exciton and valley dynamics probed by incoherent ultrafast spectroscopies are frequently perturbed due to the existence of long-lived reservoirs such as dark exciton states, leaving contradictions between the large oscillator strength and long exciton lifetime reported. Thirdly, an unambiguous extraction of the signatures of many-body interactions remains difficult for conventional spectroscopic techniques, posing an additional challenge for identifying many-body excitonic states as well as their underlying physical mechanisms.

In this perspective, we discuss how these challenges are addressed by optical two-dimensional coherent spectroscopy (2DCS) in recent experimental studies on monolayer TMDCs. We first provide a brief introduction to the 2DCS technique and several typical 2DCS configurations. Next, we highlight the capabilities of the 2DCS technique in recent milestone works on exciton coherence, valley dynamics, and exciton many-body interactions in TMDCs monolayers. Finally, we outline some potential directions for future explorations of new material systems together with the advances of 2DCS technique.

2DCS is an ultrafast nonlinear spectroscopic technique that can be viewed as an optical version of 2D nuclear magnetic resonance [3]. The idea of implementing 2DCS in the optical regime was initially proposed by Tanimura and Mukamel in 1993 [4], but it took nearly two decades to become a robust tool [5]. The major challenge for experimental realization lies in the requirement of subwavelength phase stability and interferometric precision to explicitly track the amplitude and phase of the nonlinear optical response across multiple interpulse time delays. The technique was first demonstrated using infrared ultrafast pulses [6, 7] and further extended into near-infrared and visible spectral regions with the development of phase stabilization methods [8]. By correlating quantum dynamics in two different frequency dimensions and unfolding a potentially congested 1D spectrum onto a 2D plane, 2DCS excels in measuring the optical

response of complex systems [9, 10] and has been extensively used to study energy transfer in photosynthesis [11, 12], dynamics of hydrogen bonds in water [13, 14], collective resonance in dilute atomic gas [15, 16], carrier dynamics and couplings in semiconductor quantum wells and perovskites [17–19].

Typically, 2DCS involves the measurement of the nonlinear response of a medium to a sequence of ultrafast laser pulses with tunable inter-pulse time delays [5]. Depending on the time ordering of excitation pulses and the inter-pulse delays that are scanned, there are three common types of 2D spectra [5, 8]:

(i) **Zero-quantum 2D spectrum.** In case the conjugate pulse  $A^*$  arrives first, followed by  $B$  and  $C$ , and the delays  $T$  and  $t$  are scanned, as illustrated in Fig. 1(a). Physically, pulse  $A^*$  creates a single-quantum coherence between ground and excited states, which evolves with  $\tau$ ; the second pulse  $B$  subsequently puts the single-quantum coherence into a population or a coherent superposition of the excited states during  $T$ ; the third pulse  $C$  converts the population or the superposition back to a single-quantum coherence oscillating with delay  $t$ . The generated coherence signal is heterodyne-detected and 2D Fourier transformation of the final time-domain signals yields a 2D spectrum with two frequency axes  $\omega_T$  and  $\omega_t$ . Zero-quantum 2D spectrum provides signatures about the population decay and coherent coupling between quantum states [9, 10].

(ii) **One-quantum 2D spectrum.** If the interpulse delay  $\tau$  instead of  $T$  is scanned, as depicted in Fig. 1(b), the resulting spectrum is termed as one-quantum 2D spectrum. For a three-state V system with a ground state and two singly excited states, one-quantum 2D spectrum typically features two diagonal peaks with same absorption and emission frequencies and two non-diagonal peaks with different absorption and emission frequencies due to the coupling of two transitions. One-quantum spectrum is also called rephasing or photon echo spectrum that is widely employed to investigate the

quantum decoherence process [9–12].

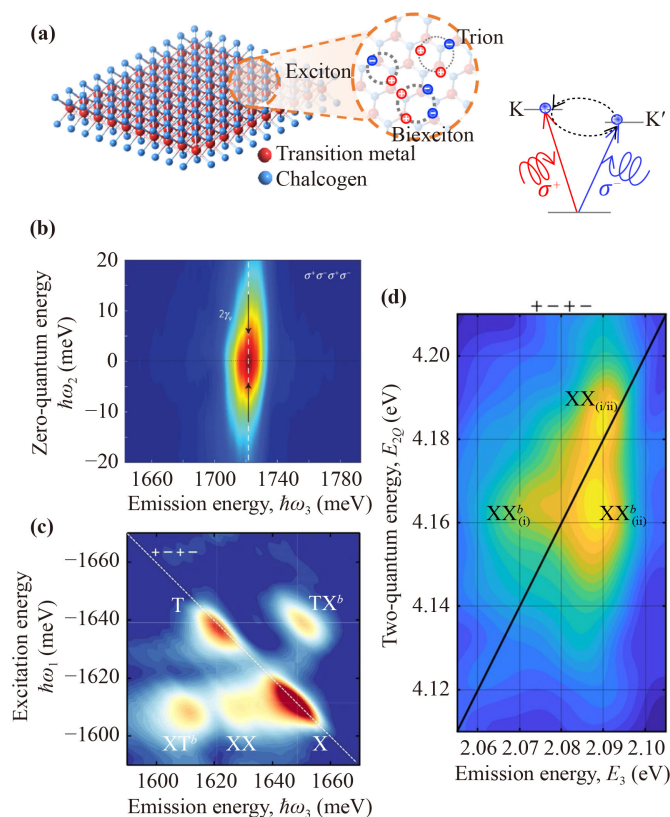
**(iii) Double-quantum 2D spectrum.** If  $A^*$  arrives last, and the delays  $T$  and  $t$  are scanned, as shown in Fig. 1(c), a double-quantum coherence between the ground state and the doubly excited state is created, resulting in a 2D spectrum with  $\omega_T = 2\omega_t$  along the diagonal line. This kind of 2D spectrum provides a sensitive and background-free detection of many-body correlations and interactions [15–17].

The unique insight provided by 2DCS on excitonic coherence dynamics in TMDCs is first demonstrated in the investigation of excitonic coherence time, which is a fundamental parameter characterizing the superposition of excitonic states and is essential for fundamental research and coherent control applications. Physically, the coherence time is related to the population relaxation time through the relation  $\frac{1}{T_2} = \frac{1}{2T_1} + \frac{1}{T_2^*}$ , where  $T_2^*$  is the pure dephasing time due to dynamical processes other than the population decay relevant to  $T_1$ . Generally, the excitonic coherence time is inversely proportional to the intrinsic linewidth of the exciton resonance. However, in an inhomogeneously broadened system like TMDCs, inhomogeneous broadening usually dominates the linewidth and conceals the homogeneous linewidth, making it a challenge to measure the intrinsic linewidth. 2DCS has the ability to overcome the ambiguities of homogeneous and inhomogeneous linewidths by projecting them into orthogonal directions in a 2D spectrum [10]. In 2015, Moody and coworkers [20] reported the first 2DCS measurement on monolayer TMDCs. In this pioneering work, the authors demonstrated that the exciton resonance of a monolayer  $\text{WSe}_2$  sample grown by chemical vapor deposition (CVD) exhibits a significant degree of inhomogeneity, obscuring the intrinsic homogeneous linewidth. By measuring the dependence of the intravalley exciton homogeneous linewidths on temperature and exciton density, an extrapolated residual homogeneous linewidth of 1.6 meV is retrieved from the one-quantum 2D spectra, corresponding to a decoherence time of  $T_2 = 0.41$  ps, which is twice the population decay time. This result indicates that the pure dephasing processes of intravalley excitons are dominated by exciton–exciton and exciton–phonon interactions. Fleming and coworkers further found that the effect of exciton–phonon coupling on the linewidth only manifested above 100 K in a CVD-grown sample of  $\text{WS}_2$  [21]. On the other hand, the  $T_2 = 2T_1$  relationship has also been confirmed in an exfoliated  $\text{MoSe}_2$  on a  $\text{Si/SiO}_2$  substrate by the Kasprzak group [22–24]. The authors integrated the collinear 2DCS technique into an optical microscope to significantly improve the spatial resolution, making the excitation region smaller than the monolayer material to accurately determine the areas yielding optical responses. Meanwhile, Cundiff and coworkers [25] developed a 2DCS scanning microscopy to map the distribution of decoherence times and inhomogeneity. In this work, a

comparative study of the neutral exciton linewidths between fully hBN-encapsulated and unencapsulated exfoliated monolayer  $\text{MoSe}_2$  showed a narrower inhomogeneous linewidth in the encapsulated samples than in the unencapsulated samples. At present, the exciton homogeneous linewidth obtained by 2DCS measurements is widely accepted as a crucial metric for assessing the quality of TMDCs materials, which is essential for researchers to optimize growth parameters and to improve the growth techniques of high-quality materials.

The quantum dynamics of excitons in TMDCs become more intriguing when considering valley physics, where excitons reside in non-degenerate valleys  $K$  and  $K'$  with opposite electronic spin. The observation of valley physics was enabled by the valley-dependent optical circular dichroism, where the excitonic states in the  $K(K')$  valley can be excited by  $\sigma^+$  ( $\sigma^-$ ) circularly polarized light [26, 27], offering a proper tool for valley pseudospin manipulation. To achieve quantum control of the valley pseudospin, it is crucial to measure and control not only the valley population but also the valley coherence dynamics. However, the detection of the valley coherence dynamics is challenging for linear spectroscopy because exciton valley coherence is a classic example of non-radiative coherence for which exciton transitions are parity-forbidden. Zero-quantum 2D spectrum provides an effective tool for probing the coherence between quantum states that are not dipole-coupled. By analyzing the slice along the mixing frequency axis in the zero-quantum 2D spectrum using cross-circular polarization, as shown in Fig. 2(b), it has been possible to directly extract an exciton valley coherence time of approximately 100 fs in a monolayer of  $\text{WSe}_2$  [28].

The neutral excitons can trap an extra electron or hole to form trions, which significantly influence the physical properties and bring in new physics to the quantum dynamics of TMDCs materials. 2DCS measurements on CVD-grown  $\text{MoSe}_2$  have revealed an intrinsic homogeneous linewidth of trion resonance with a coherence dephasing time of  $T_2 = 182$  fs [29]. Similar to exciton valley coherence, trion valley coherence time of about 230 fs in  $\text{MoSe}_2$  monolayer has been detected using the zero-quantum 2D spectrum [30]. It is also found that the valley coherence time of excitons is governed by the population lifetime, while the valley coherence time of trions is dominated by the pure dephasing caused by the properties of samples, such as impurities, defects, substrate, strain, and carrier density. Particularly, as carrier density increases, the trion picture falls short in explaining emerging phenomena in materials [31, 32]. The concept of a Fermi polaron, which describes an exciton dressed by its surrounding Fermi sea, has been proposed. Recent 2DCS studies have revealed interactions between Fermi polarons and bound bipolarons in monolayer TMDCs [31]. Distinct properties of attractive and repulsive polarons were



**Fig. 2** (a) Left: Monolayer TMDCs crystal structure, along with the excitons and excitonic complexes. Right: Exciton resonance with valley-dependent optical selection excitation. (b) Zero-quantum 2D spectrum of monolayer WSe<sub>2</sub> obtained with the cross-circular polarization pulses. The linewidth extracted along the mixing frequency axis gives the valley coherence time. (c) One-quantum 2D spectrum of monolayer MoSe<sub>2</sub> obtained using cross-circular polarization pulses. The peaks X and T denote the excitons and trions, and the additional peaks XX and TX<sup>b</sup> (XT<sup>b</sup>) are associated with the neutral and charged bound biexciton, respectively. (d) Double-quantum 2D spectrum shows an unbound two-excitons state XX and bound biexciton state XX<sup>b</sup>. Panels adapted with permission from: (b) Ref. [28]; (c) Ref. [43], under a Creative Commons license CC BY 4.0; (d) Ref. [45].

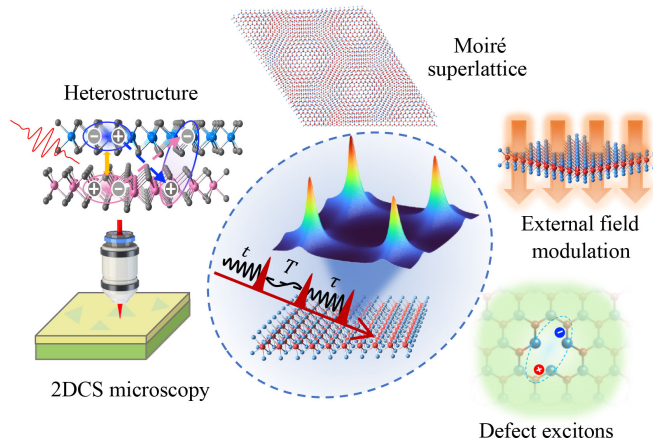
observed as the carrier density varies. More specifically, the dephasing rate of repulsive polarons increases quadratically while attractive polarons remain stable [32].

Phonon effect in TMDCs is found to be critical in determining the quantum dynamics of excitons and their derivatives [21, 29, 33–35]. The one-quantum 2D spectrum of monolayer MoSe<sub>2</sub> revealed a larger Huang–Rhys factor compared to most inorganic semiconductor nanostructures at room temperature [34]. Furthermore, by embedding TMDCs in a microcavity, the coupling of exciton and cavity photon forms the exciton polaritons and multiple polariton branches induced by exciton–photon–phonon hybridization were observed in one-quantum 2D spectrum

[35]. These results are important for realizing high-speed and low-loss optoelectronic devices at room temperature.

Another far-reaching advantage of 2DCS is its unique insights into the excitonic many-body coupling, making critical and timely contributions towards a fully comprehension of the exciton physics in TMDCs materials. In semiconductors, excitons can interact with each other and with free carriers, leading to significant many-body effects. Due to their band structure, TMDCs materials typically present two species of excitons (A and B excitons) with different resonance energies. In MoS<sub>2</sub>, the intravalley exchange interaction between A and B excitons can be observed as cross-peaks in one-quantum 2D spectrum [36, 37] and it was found that the intervalley coupling dynamics do not depend on temperature and carrier density. Moreover, the 2D spectrum has shown that the in-plane magnetic field brightened dark excitons are coherently coupled with bright excitons [38]. Additionally, a similar coupling between excitons and trions was also revealed in monolayer TMDCs [39–42]. The coherent coupling between excitons and trions in monolayer TMDCs was first reported using two-color pump-probe 2D spectroscopy [39]. Isolated cross-peaks originating from the coupling between excitons and trions appear in the spectra, and the spectral lineshape reveals the excitation-induced shift and excitation-induced dephasing many-body effects. However, this method is insufficient in unraveling comprehensive many-body coupling dynamics. In contrast, one-quantum 2DCS has complementarily revealed different coupling dynamic processes at different timescales [40]. Coherent coupling between excitons and trions governs the dynamics on a timescale of hundreds of femtoseconds, while phonon-assisted exciton-to-trion down-conversion and trion-to-exciton up-conversion processes dominate the coupling over tens of picoseconds. Recently, the manipulation of the coupling strength between excitons and trions has been demonstrated using a gate voltage, offering new possibilities for developing novel optoelectronic devices and quantum information technologies [41].

In addition to the interaction between neutral and charged excitons, the Coulomb force can bind multiple particles into high-order tight-bound states, such as biexciton states. However, the identification of the biexcitons from 1D spectrum measurements is extremely challenging due to confusion arising from other quasiparticles, such as electron plasma in TMDCs. 2DCS provides conclusive evidence of the existence of biexcitons by expanding the convoluted 1D spectrum into 2D [43–45]. On one hand, a comparison of co- and cross-circularly polarized one-quantum 2D spectra unambiguously revealed the neutral and charged bound biexcitons with binding energies of 20 and 5 meV, respectively [43], resolving the debate between the measured and calculated neutral biexciton binding energy [46, 47]. Theoretical simulations further identified that these biexcitons are



**Fig. 3** Future directions of 2DCS in TMDCs.

composed of excitons or trions residing in opposite valleys, which can be used to generate entangled photons. On the other hand, the double-quantum 2D spectrum provides an alternative approach for the direct detection of biexcitons by measuring the double-quantum coherence between the ground state and the doubly-excited state of biexcitons. Peaks  $XX$  and  $XX^b$ , arising from the unbound two excitons and bound biexciton, as shown in Fig. 1(c), were readily observed in the spectrum [45].

2DCS has been demonstrated as a powerful tool for investigating exciton physics in TMDCs. Despite significant progresses that have been achieved, there are still many exciting opportunities to be explored in this field, boosted by the most recent advancements in 2DCS techniques and new materials. Below, we have compiled a list of possible representative directions as depicted in Fig. 3.

**2DCS microscopy.** Traditional 2DCS techniques are limited to specific points on the sample owing to their low spatial resolution and slow data acquisition speed. However, recent technical developments in colinear 2DCS microscopy have opened up new avenues for investigating the optical response of each point and mapping the inhomogeneity of TMDCs with optical diffraction-limited spatial resolution [22, 25]. By introducing non-optical detection methods such as photoemission electron microscopy, spatial resolution beyond the optical diffraction limit can be achieved in 2DCS microscopy [48]. These techniques offer numerous possibilities, including scanning low-strain areas for quantum information applications, guiding the manufacture of materials for specific purposes, characterizing defects, and imaging nanoscale surface coherence. They can also be used to study bilayer structures and identify the distribution of moiré excitons, which are becoming increasingly important for gaining deeper insight into condensed matter physics and developing novel optoelectronic devices.

**Heterostructures.** TMDCs offer a promising platform for creating van der Waals heterostructures with diverse properties [49]. Typically, TMDCs heterostructures have a type-II band alignment, where electrons and holes find their energy minima at different layers, enabling the formation of interlayer excitons. Recent 2DCS studies have explored the exciton coherent coupling and binding energy of tightly bound interlayer excitons in  $\text{MoSe}_2/\text{WSe}_2$  heterostructures, as well as interlayer hole–electron transfer in  $\text{WS}_2/\text{MoS}_2$  heterostructures [50, 51]. However, many aspects of interlayer excitons, including their coherent coupling, their interaction with intralayer excitons, charge transfer, energy transfer, and dependence on interlayer twist angle, remain exciting opportunities for future research.

**Moiré superlattices.** As a specialized 2D heterostructure, moiré superlattices, which is formed by lattice mismatch or twist angle in vertically stacked TMDCs materials, is one of the most popular topics in the field of 2D materials. Due to TMDCs materials' non-degenerate energy band structure, there is a large twist angle range to find flat Moiré mini bands, deep moiré periodic potential, and consequently spatial localization of electrons compared with twisted bilayer graphene [52, 53]. In TMDCs moiré superlattices, excitons from multiple moiré mini bands induce the appearance of moiré excitons, dominating the optical properties of TMDCs moiré superlattices. However, a fundamental question is left open until now: how do the moiré superlattices influence the quantum dynamics of moiré excitons? Moreover, considering excitons are widely applied to probe the abundant correlated electronic phases in TMDCs-based Moiré superlattices, this raises another intriguing question: how do these correlated electronic phases influence excitonic quantum dynamics? Overall, the vast majority of quantum dynamics in moiré superlattices remains enigmatic, leaving abundant opportunities for 2DCS.

**External field modulation.** One of the exceptional features of atomically thin 2D materials is easy modulation by the external fields owing to their low dimensionality. The electric field is widely applied to modify the Fermi level of TMDCs materials and bring in an electric field effect, which induces a plethora of fascinating phenomena, such as Fermi polaron formation, Stark effect, and the modified coupling strength between quasiparticles. Similarly, the magnetic field, as another commonly used tuning knob, can create Landau levels when applied to TMDCs materials in out-of-plane configuration, which consequently modifies excitons and enhances higher-order four-particle correlations [54, 55]. In addition, in-plane magnetic fields can brighten the optically forbidden “dark” excitons and facilitate their interaction with bright excitons. There are far more species of fields that can significantly impact TMDCs materials, e.g., strain field, optical field, and pseudo magnetic field. 2DCS holds tremendous potential for investigating these inter-

esting properties induced by external fields and for unveiling their underlying mechanisms.

**Defect excitons.** Defects in low-dimensional materials give rise to a plethora of intriguing phenomena, especially the emergence of single photon emitters (SPEs) [56]. Since their discovery in 2015, the ultrasharp single photon emission lines observed in TMDCs have attracted significant attention, due to their deep insights into associated SPE physics and potential applications in quantum information. Despite the remarkable advancement achieved since then, many challenges still lie ahead. Under intensive investigation, the origin of SPEs in 2D materials remains elusive as a fundamental question in both science and applications, which is inferred as related to the localized excitons or trions confined by the local fields created by point defects or local strains. Moreover, considering the diverse array of defects in 2D materials, such as point defects, line defects, and grain boundaries, the intricate interactions between defects, excitons, and other quasiparticles call for further investigation. In this context, we anticipate 2DCS will shed light on the investigation of defect-related phenomena in 2D materials, potentially enhancing our insights and revealing unexplored physical processes and properties therein, including the physical origin of SPEs, defect-induced strong correlation, etc.

**Declarations** The authors declare that they have no competing interests and there are no conflicts.

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