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Gate tunable dark trions in monolayer WSe_2

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Abstract: Monolayer WSe_2 is an intriguing material to explore dark exciton physics. We have measured the photoluminescence from dark exciton and trions in ultraclean WSe_2 devices encapsulated by boron nitride. The dark trions can be tuned continuously between negative and positive trions with electrostatic gating. We reveal their spin triplet configuration and distinct valley optical emission by their characteristic Zeeman splitting under magnetic field. The dark trion binding energies are 14 – 16 meV, slightly lower than the bright trion binding energies (21 – 35 meV). The dark trion lifetime (~1.3 ns) is two orders of magnitude longer than the bright trion lifetime (~10 ps) and can be tuned between 0.4 and 1.3 ns by gating. Such robust, optically detectable, and gate tunable dark trions may help us realize trion transport in two-dimensional materials.

Monolayer transition metal dichalcogenides (TMDs), such as MoS_2 and WSe_2, are remarkable two-dimensional semiconductors with strong Coulomb interactions. Their optical properties are governed by tightly bound excitons across the direct band gaps in two time-reversal valleys (K, K') [1-4]. The strong spin-orbit coupling splits both the conduction and valence bands into two subbands with opposite spins [Fig. 1] [5-8]. The spin configuration governs an exciton's optical properties. If the electron and hole come from bands with the same electron spin, their recombination can efficiently emit light. These bright excitons have short lifetime (<10 ps) [9], in-plane transition dipole moment and out-of-plane light emission [Fig. 1] [10]. But if the electron and hole come from bands with opposite electron spins, the spin mismatch strongly suppresses their radiative recombination. They form dark excitons with long lifetime (>100 ps), out-of-plane transition dipole moment, and in-plane light emission [Fig. 1] [11-19].

Monolayer WSe_2 is an excellent material to explore dark exciton physics. Its dark exciton level lies well below the bright exciton level [Fig. 1] [7, 12, 20]. The dark excitons can thus accumulate a large population to achieve observable light emission [17, 21-24]. We can control the charge density of the material by using electrostatic gating. In finite charge density, dark excitons can interact with the Fermi sea to form dark trions [Fig. 1] [25] (here we use "trions" to broadly refer to any bound states between exciton and an excitation from Fermi sea). The dark trion states are intriguing entities for research and applications, but measuring them is challenging due to their low optical activity. Recent research has revealed the evidence of dark trions in monolayer WSe_2 under strong in-plane magnetic field [15] and plasmonic environment [16], but there is still a lack of investigations on their detailed gate-dependent properties.

In this letter, we measure the photoluminescence (PL) of dark trions in ultraclean monolayer WSe_2 devices encapsulated by boron nitride (BN). The supreme quality of our devices allows us to observe both the positive and negative dark trions under continuous electrostatic gating. Compared to bright trions with binding energies 21 – 35 meV, dark trions have smaller, but still sizable, binding energies (14 – 16 meV). We reveal the spin triplet configuration and distinct valley optical emission of dark trions by their characteristic Zeeman splitting under magnetic field. Their g-factors (~ –9) are about twice of that of bright trions (~ –4) with spin singlet. Notably, the dark trion lifetime (1.3 ns) is two orders of magnitude longer than the bright trion lifetime (~10 ps). The dark trion lifetime can be tuned continuously between 0.4 and 1.3 ns by gating. Such robust, optically detectable, and gate tunable dark trions provide us with an excellent platform to explore novel excitonic physics.

We measured the PL of monolayer WSe_2 with 532-nm continuous laser excitation at temperature T ~ 4 K. We applied very low incident laser power (16 μW) to suppress the biexciton PL, which is known to be significant in monolayer WSe_2 [21-24]. Our samples emit weak but noticeable PL from dark exciton/trions. Although such PL propagates in the in-plane direction, we can partially capture it with a wide-angle microscope objective (NA = 0.67) in the out-of-plane detection geometry [17]. Compared to previous detection methods that require either in-plane magnetic field, plasmonic coupling, or near-field tip enhancement [13-16], our detection method is much simpler and preserves the intrinsic properties of dark excitonic states.

Fig. 2(a) displays a gate-dependent PL map of monolayer WSe_2. We observe the bright A exciton (A^0) at 1.712 meV at the charge neutrality point (CNP) (gate voltage V_g = –0.5 V). On the electron and hole sides, one positive (A^+) and two negative (A_1^-, A_2^-) bright trions appear. Their binding energies are 21, 29 and 35 meV, respectively, consistent with prior reports [26, 27]. At the charge neutrality point, a weak PL peak appears at 41 meV below the bright exciton. This is the dark exciton (D^0) according to prior studies [14-17, 21-24]. As we tune WSe_2 to the electron and hole sides, the D^0 peak subsides and two new peaks emerge at 16 and 14 meV below the D^0 peak. We denote them as D^- and D^+, respectively. Both the D^- and D^+ intensity increases linearly with the excitation laser power; they are thus not associated with biexcitons with quadratic power dependence [28]. We have multiple experimental evidence to support that the D^- and D^+ features come from the dark trions.

First, the D^0, D^- and D^+ features exhibit parallel gate dependence to the bright exciton/trions. We compare the gate-dependent PL energy, intensity and line width of the D^0, D^-, D^+ peaks with those of bright exciton/trions [Fig. 2(b-d)]. They exhibit parallel gate-dependent behavior. This suggests that the D^- and D^+ peaks correspond to the negative and positive dark trions, respectively. Moreover, the energy separation (14 – 16 meV) between the D^-, D^+ peaks and the D^0 peak matches the reported binding energy [15, 28] and the theoretically predicted binding energy (~14 meV) [36] of dark trions in monolayer WSe_2.

Second, they exhibit opposite temperature dependence from the bright exciton/trions. When we decrease the temperature, the bright trion PL drops continuously, but the dark trion PL grows stronger (see Fig. S8 in the Supplementary Materials [28]). Such contrasting behavior reflects their different energy levels and population distribution. The bright trions lie at higher energy and their population drops with decreasing temperature, but the dark trions lie at the lowest energy and their population increases at low temperature [Fig. 1] [13].

Third, dark trions have a distinct spin configuration from that of bright trions. While bright trions involve a spin-singlet exciton, dark trions involve a spin-triplet exciton (see Fig. 1; the hole has the opposite spin of the valence electron) [18]. These different spin configurations can be distinguished by Zeeman effect.

Fourth, the dark trions follow different optical selection rules from those of bright trions. According to prior research, bright excitons at the K (K') valley emit light with right-handed (left-handed) circular polarization in the out-of-plane direction [6, 37, 38], but dark excitons at both K and K' valleys emit light with vertical linear polarization in the in-plane direction [Fig. 1] [17]. Their associated bright/dark trions follow the same optical selection rules, because the recombination of exciton is only weakly affected by its coupling to the Fermi sea [39].

We can confirm the third and fourth characteristics by PL measurements under out-of-plane magnetic field, which lifts the valley degeneracy in monolayer TMDs [29, 32, 34, 35]. Due to the opposite spin and orbital configurations of the K and K' valleys, the magnetic field can enlarge the band gap of one valley and diminish the band gap of the other valley [Fig. 3(a)]. The difference between the two valley gaps is defined as the valley Zeeman splitting energy $\Delta E = g\mu_{\mathrm{B}}B$, where g is the effective g-factor and $\mu_{\mathrm{B}} = 57.88\,\mu eV/T$ is the Bohr magneton. For bright exciton/trions, their Zeeman splitting has been argued to mainly come from the orbital magnetic moment of the valance band, with an estimated g-factor of about –4 [29, 32, 34, 35]. But for dark exciton/trions, their associated spin triplet will contribute an additional g-factor of –4, making their total g-factor about –8 (see Supplementary Material for more discussion) [18, 28, 33].

To further probe the distinct optical selection rules of dark and bright trions, we adopt a special measurement geometry – we excite monolayer WSe_2 with unpolarized light but collect only the right-handed PL. For bright exciton/trions with circularly polarized PL, our measurement only detects the light emission from the K valley. But for dark exciton/trions with linearly polarized PL, our measurement detects emission from both valleys.

Figure 3(b-d) display the gate-dependent PL map at B = –10 T and the B-dependent PL maps from B = –31 to 31 T. The spectra of bright exciton/trions remain largely unchanged except for a Zeeman shift – we detect light from only one valley as expected. Their g-factors (–4.1 to –4.4) also matches our prediction (–4) from their atomic orbits and spin singlet configuration [Fig. 3(e)] [29, 32, 34, 35]. In contrast, the D^0, D^-, D^+ peaks each split into two peaks with the same energy separation – we detect light emission from both valleys as expected from dark exciton/trions. Moreover, their g-factors (–9.3 to –9.5) roughly match our prediction (–8) from the atomic orbits and spin triplet configuration of the dark states [Fig. 3(e)] [18, 28, 33]. Our experiment therefore confirms both the spin triplet configuration and the distinct valley optical selection rules of the dark trions in monolayer WSe_2.

Dark trions are expected to live much longer than bright trions, because their spin triplet configuration suppresses the radiative decay [15, 18]. The longer lifetime of dark trions is hinted from their narrower line widths (FWHM ~ 2.5 meV) compared to bright trions (FWHM = 3 – 9 meV) [Fig. 2(d)]. We have measured the time-resolved PL intensity of trions by the time-correlated single photon counting method [Fig. 4(a)] [28]. The bright trion PL dynamics exhibits two exponential decay components. The fast and dominant component follows closely the instrument response function (IRF). We extract a lifetime of ~10 ps after deconvolution with the IRF. The slow component contributes little because it is much weaker than the fast component [inset of Fig. (4a)] [40-42]. In contrast, the dark trion (at V_g = –0.8 V) shows a single exponential decay with a lifetime of 1270 ps. It is two orders of magnitude longer than the bright exciton/trion lifetime (1~10 ps) and an order of magnitude longer than the dark exciton lifetime (180 ps) [Fig. S12] [18, 28].

The lifetime of dark trions is gate tunable and correlates with the PL intensity and line width. Fig. 4(b) displays the dark trion lifetime, PL intensity and line width as a function of gate voltage (V_g). When V_g changes from 0 to –1 V in the hole side, the dark trion lifetime increases slightly from 1 to 1.3 ns; the PL intensity rises; the line width remain almost constant. When V_g goes further to –4.4 V, the dark trion lifetime decreases from 1.3 ns to 370 ps; the PL intensity decreases; the line width increases correspondingly [28]. These observations can be qualitatively understood from the interactions between trions and free carriers. When carriers are first injected into the sample, they facilitate the trion formation and enhance the trion PL. But as their density increases, they will scatter frequently with the trions. Such scattering will shorten the trion lifetime, suppress the trion PL, and broaden the PL peak.

Prior research has shown that trions can behave like free carriers with controlled motion under electric field [43, 44]. Their charge, spin, valley and internal energy degrees of freedom make them attractive information carriers. But trion transport has not been achieved in atomically thin materials. One primary reason is the short lifetime of bright trions. The long-lived dark trions offer an effective solution. We may assume the trion mobility (μ) to be one-tenth of the electron/hole mobility because trions have triple effective mass and higher scattering rate. The trion mobility can reach μ = 400 cm^{2}V^{-1}s^{-1} from the highest carrier mobility (4,000 cm^2 V^{-1} s^{-1}) reported in monolayer WSe_2 [45]. By using an electric field $E = 0.5\,\mathrm{V/\mu m}$ and our observed lifetime $\tau = 1\,ns$, the drift distance of dark trions under in-plane electric field can reach $l_d = \mu E \tau = 20\,\mu m$. This distance exceeds the typical length scale of two-dimensional material devices. The long-lived, optically detectable and gate tunable dark trions therefore provide a new path to investigate field-controlled trion transport.

Finally, we note that some researchers interpret the excitonic states under doping as exciton polarons (not trions) [46, 47]. In this picture, an exciton evolves from a trion-hole state in low doping to an exciton polaron in high doping [48]. Our experimental conclusion is still valid in this picture by simply changing “dark trions” into “dark exciton polarons”. The trion transport should still be realizable in the low doping regime, where the loosely bound trion-hole states easily dissociate into a trion and a free hole under in-plane electric field.

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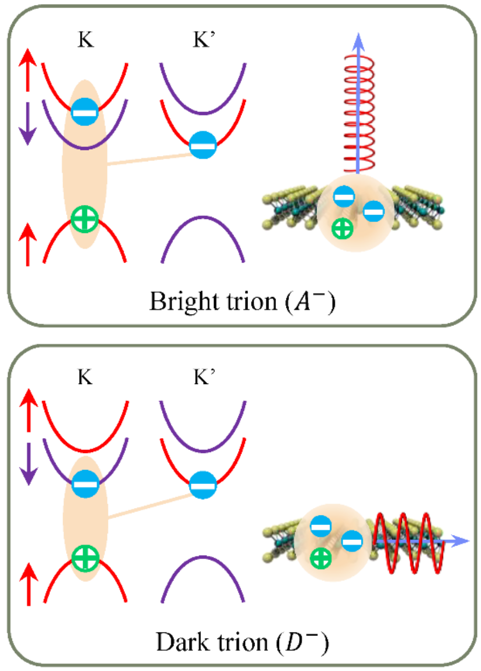

FIG. 1. Intervalley bright and dark excitons/trions in monolayer WSe_2. The red (blue) lines denote bands with up (down) electron spin. The shaded ellipses denote excitons in the recombination. The bright exciton/trions emit circularly polarized light in the out-of-plane direction. The dark exciton/trions emit vertically polarized light in the in-plane direction.

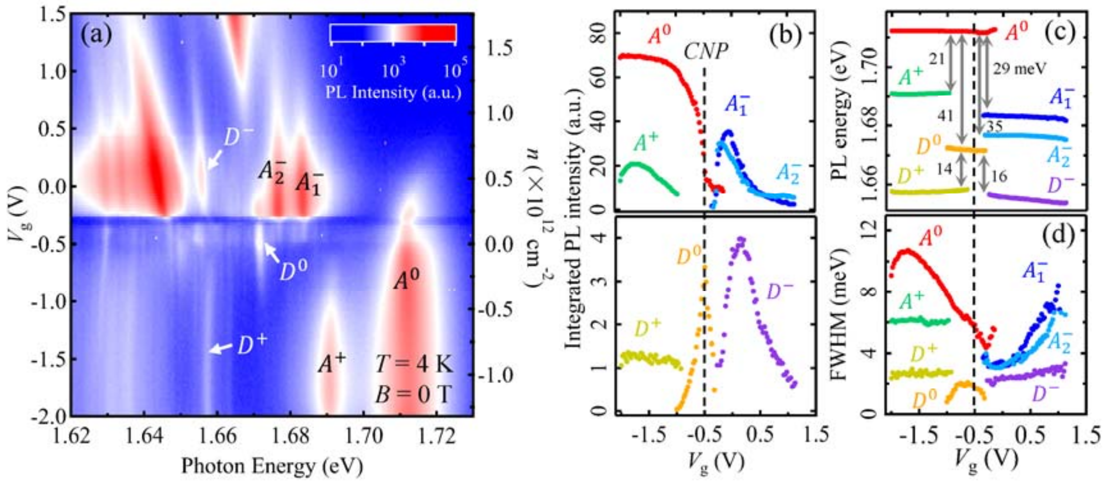

FIG. 2. (a) Gate-dependent PL map of BN-encapsulated monolayer WSe_2 (Device 1) at T = 4 K and B = 0 T under 532-nm continuous laser excitation. (b) The extracted PL integrated intensity, (c) PL energy, (d) PL full width at half maximum (FWHM) of dark/bright excitons/trions as a function of gate voltage. The numbers in panel (c) denote the energy separation in unit of meV. The dashed lines denote the charge neutrality point (CNP).

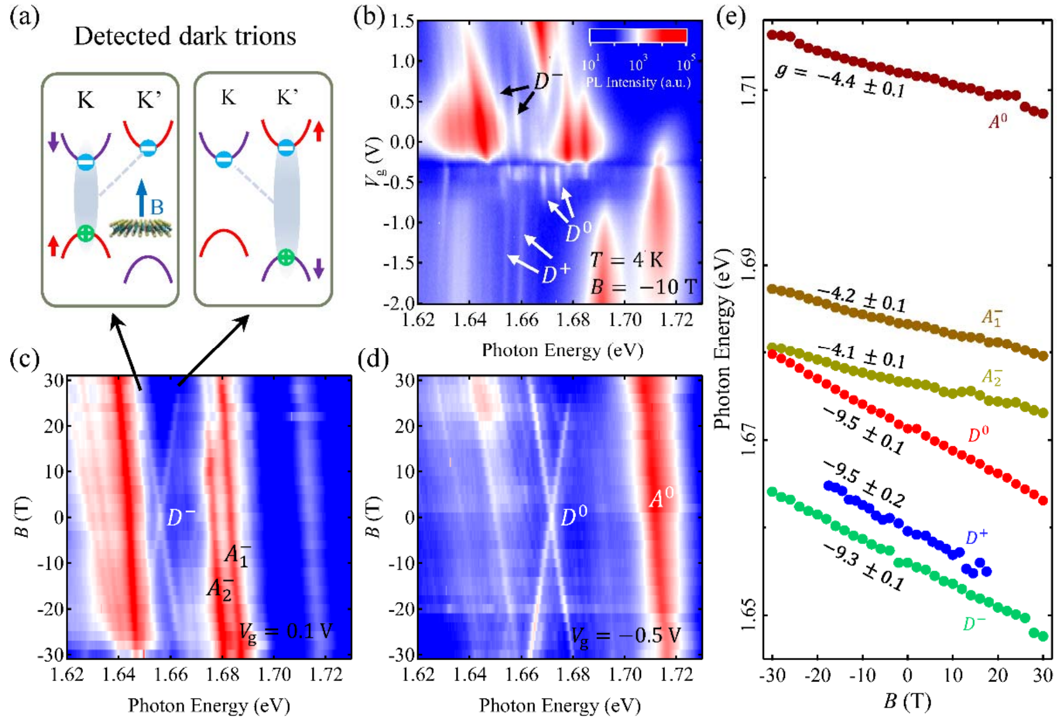

FIG. 3. (a) Negative dark trions detected in our magneto-PL experiment. (b) The PL map of monolayer WSe_2 (Device 1) at $B = -10$ T under unpolarized optical excitation and right-handed PL detection. Dark exciton/trions are split into two peaks, whereas bright exciton/trions are not. (c-d) The B-dependent PL map at the electron side (V_g = 0.1 V) and charge neutrality point (V_g = –0.5 V). Panels (b-d) share the same color scale bar. (e) The exciton/trion Zeeman shifts. The g-factors are extracted by linear fits of the energy difference between positive and negative magnetic field.

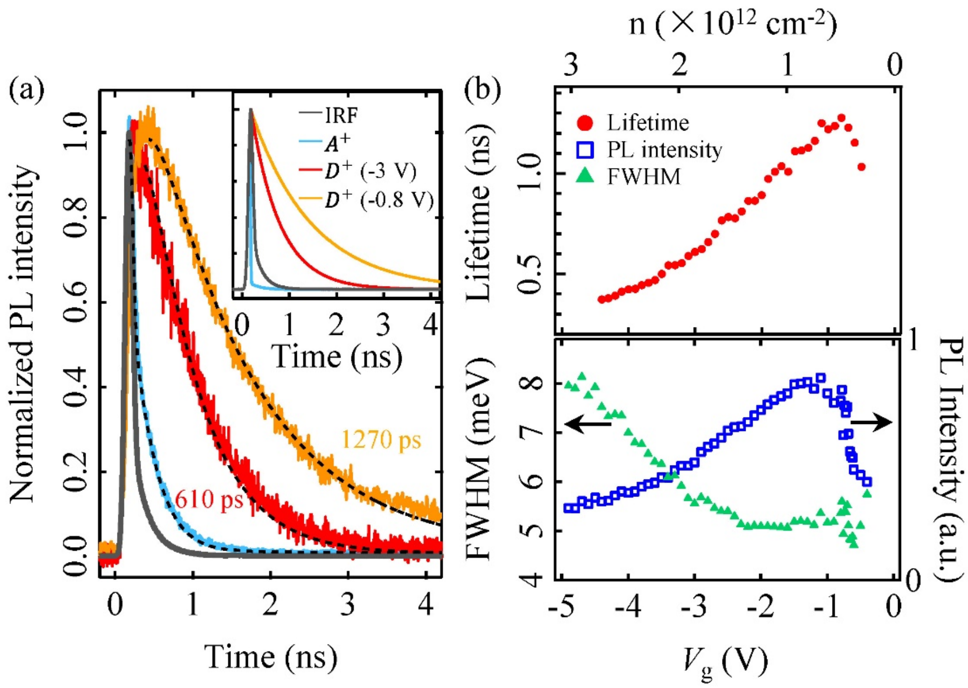

FIG. 4. (a) Time-resolved PL for the A^+ bright trion and D^+ dark trion in BN-encapsulated monolayer WSe_2 (Device 2). The black solid line is the instrument response function (IRF). We fit the D^+ and A^+ data, respectively, with single-exponential and biexponential functions convolved with the IRF (dashed lines). The inset shows the deconvolved fits. The D^+ lifetime is 1270 ps and 610 ps at gate voltages V_g = –0.8 V and –3 V, respectively. The A^+ lifetime is ~10 ps. (b) The lifetime, PL intensity and full width at half maximum (FWHM) of the D^+ trion as a function of gate voltage.