Exciton emissions of bilayer WSe₂ tuned by the ferroelectric P (VDF-TrFE) polymer

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Abstract
In this work, we show electrical polarization of ferroelectric P (VDF-TrFE) polymer can be used to engineer the photoluminescence (PL) at bilayer WSe₂. The total PL intensity substantially is suppressed under negative polarization and enhanced in positive polarization with increasing the polarization intensity of P (VDF-TrFE) polymer. And the electron transfer between conduction band energy valleys K and Λ due to built-in electric field can modify the recombination path and change the overall optical radiation efficiency, and analysis of the change is performed by the rate equation based on the charge transfer theory. The calculated intensities well reproduce the experimental results. This work may create an opportunity for hybrid integration of ferroelectric materials and 2D TMDCs as optical switch.

Keywords: WSe₂, P (VDF-TrFE), photoluminescence, charge transfer

Introduction
Thin layered transition dichalcogenides (TMDCs) due to high area-to-volume ratio, atomically thin are considered a promising materials for optoelectronic devices.¹⁰⁻¹¹ TMDCs have unique electrical and optical characteristics, evolving from an indirect band gap to a direct bandgap when the number of layers is reduced to a single layer.¹²⁻¹⁴ Besides fundamental studies including bound excitonic states,¹⁵⁻¹⁷ valley polarization,¹⁸⁻²⁰ the 2D TMDCs have been used in photodetectors,²¹⁻²⁷ nonlinear optical devices and frequency converters,²⁸⁻³⁰ photovoltaic³¹,³² and advanced non-volatile memory³³,³⁴. Due to the ultra-thin nature of TMDCs, few layer TMDCs materials have a significant response to the surrounding environment. Several external tuning approaches such as temperature,³⁵⁻³⁷ strain,³⁸⁻⁴¹ chemical doping,⁴²⁻⁴⁵ have been demonstrated to alter the electronic and optical properties of few layer TMDCs. Moreover, this opens up an opportunity for ferroelectric gate to adjust the properties of few layers of TMDCs.²⁷,⁴⁶,⁴⁷ Ferroelectric materials possess a spontaneous electrical polarization resulting in a strong built-in electric field. The electrical polarization can be control through application of an external voltage. P (VDF-TrFE) polymer is a widely known ferroelectric material for piezoelectric sensors, photodetectors.²⁷ Based on its excellent
transparency from visible to infrared wavelengths, it is feasible to explore the optical response based on hybrid integration of P (VDF-TrFE) and 2D TMDCs. Recently, WenBo et al. reported that polarized lithium niobate can modulate the exciton and trion in monolayer MoSe$_2$ and WSe$_2$.

Moreover, 2D van der Waals-ferroelectric heterostructures present a tantalizing opportunity to complete the integrated functional components on a single chip. Compared with monolayer TMDCs, exploring the integration of multilayer TMDCs thin film and ferroelectric materials has far-reaching significance in photonics due to high electron mobility and larger optical density of state in multilayer TMDCs. However, until now, there is basically no corresponding report on the use of ferroelectric materials to adjust the optical characteristics of multilayer TMDCs.

Herein, we explore the interaction of polarized P (VDF-TrFE) thin film with bilayer WSe$_2$ and observed opposite modulation of the PL intensity. The electrical polarization of P (VDF-TrFE) polymer is adjusted through application of an external gate voltage. The experimental observed PL intensity is substantially suppressed in negative polarization and enhanced in positive polarization with increasing the gate voltage strength. Strain caused by the inverse piezoelectric effect, and thermal effect are not the main factors that cause the PL intensity to change. We suggests that the observed PL changes are due to the transfer of intervally electrons between conduction band energy valleys K and Λ, and readjust the recombination path. The calculated intensities well reproduce the experimental results. We believe that these results may be open a new avenue for optical switch based on the hybrid integration of ferroelectric materials and 2D TMDCs.

**Experimental methods**

The bilayer WSe$_2$ samples were mechanically exfoliated from bulk WSe$_2$ onto SiO$_2$/Si substrates. The PL signals excited by a 514.5 nm line of an Ar-ion laser were collected by tri-vista Raman spectroscopy equipped with a liquid nitrogen-cooled Si-CCD camera. Relatively low excitation power is used to reduce the effect of thermal effect.

The fabrication process of the entire test structure can be find from the reference. Corresponding electrical characteristics are shown in the supplemental materials. After measuring the back bias, the sample is annealed at 400 °C to eliminate the residual polarization of P (VDF-TrFE), and then the test under forward bias. The ferroelectricity of P (VDF-TrFE) film is stable at room temperature, so we first study the control characteristics of P (VDF-TrFE) thin film at room temperature.

The electronic structures of bilayer WSe$_2$ were calculated through the density functional theory (DFT) code VASP. In our calculation, the exchange correlation potential was dealt with hybrid functionals at both Perdew–Burke–Ernzerhof (PBE) levels, and the cutoff energy was set to 450 eV. A vacuum of 20 Å perpendicular to the surface was used to demonstrate finite layer, and a 15 × 15 k-meshed grid was employed to describe the periodic properties of bilayer WSe$_2$. The convergence criterion was restricted to less than 10$^{-5}$ eV in energy. The atomic positions were fully relaxed until the Hellmann–Feynman forces acting on each atom were less than 0.01 eV Å$^{-1}$.

**Results and discussion**

Figure 1(a) displays the 3D schematic diagram of the two-terminal device configuration after coating with P (VDF-TrFE). The source and drain electrodes are on the WSe$_2$ flake, and Pd/Au is selected as a contact electrode to improve the contact resistance. The hysteresis loop of 300 nm P(VDF-TrFE) at room temperature is shown in figure 1(b). When the electric field strength
increases to near ±60 V, it tends to be in a polarization saturation state. At this time, the electric field strength further increases, the total polarization strength will still gradually increase as the induced polarization increases. The coercive voltage is approximately 22.5 V and the remnant polarization value is 7 μC/cm². Figure 1(c) show the transfer characteristics of the bilayer WSe₂ transistor at 300K. The PL spectra for bilayer WSe₂ after coating with P (VDF-TrFE) at T=300 K are presented in figure 1(d). A high energy PL peak (A) at 1.58 eV originating from the direct interband transition K→K. Additional a peak (I) emerges from the indirect bandgap emission K→Λ. The measured thickness of bilayer WSe₂ is about 1.5 nm, matching the PL peak of the measurement.

Figure 1. (a) The 3D schematic diagram of the two-terminal device configuration after coating with P (VDF-TrFE). The Pd/Au serves as the drain and source electrodes and Al serves as the top-gate electrode. (b) The P (VDF-TrFE) polarization-voltage curves at T=300 K. (c) Transfer curve of the bilayer WSe₂ transistor at T=300 K. (d) Bilayer WSe₂ PL spectra at room temperature. The blue and green lines indicate the direct (A) and indirect (I) interband transitions, the dashed lines are only added as an eye guide.

The behavior of bilayer WSe2 under a negative vertical electric field is completely different than the positive case, with the total PL enhanced, as illustrated in the PL spectra in figure 2. We have repeated these measurements on more than 4 times to confirm the consistency of the behavior. In addition to the intensity change, the vertical electric field also cause the PL peak to red-shift slightly. Inset in figure 2 show the total integrated PL intensity versus gate voltage. As the field strengthens to a certain threshold value (~ -60 V), the total PL signal increases slowly. This spectral behavior is consistent with the hysteresis loop change trend. By proper control of experimental conditions, the PL enhancement could reach approximately 3 times. The positive vertical electric field condition can be found from the supplementary figure S1.
Figure 2. PL spectra at negative gate voltage with bilayer WSe$_2$. The change of PL intensities with the increase of the applied bias is shown in the inset. The dashed lines are only added as an eye guide.

In order to analyze the intensity change mechanism. As a comparison, we tested the PL spectrum with bilayer WSe$_2$ under different applied electric fields in the case of back gate structure (SiO$_2$ dielectric). As shown in figure S2, Directly applied bias voltage on bilayer WSe$_2$ has no modulation effect on the PL intensity of bilayer WSe$_2$. From the above discussion, it can be concluded that the change in PL intensity is due to the effect of ferroelectric P(VDF-TrFE) crystals.

This may be related to the deformation of P (VDF-TrFE) thin film based on the inverse piezoelectric effect. Reverse bias causes the P (VDF-TrFE) thin film to stretch or shrink, leading to the bilayer WSe$_2$ to deform. To our knowledge, this effect is not obvious in the polymer ferroelectric materials. In addition, the deformation of P (VDF-TrFE) disappears with the removal of the applied gate voltage. In order to evaluate the effect of strain, the PL measurements under different levels of remnant polarization with P (VDF-TrFE) are recorded. The applied negative gate voltage is removed after polarizing P (VDF-TrFE). As can be seen in figure 3, total PL intensity increases as residual polarization increases, and the intensity is basically unchanged after reaching a critical value ($7 \mu$C/cm$^2$ or above -60 V). This suggests that deformation introduced by inverse piezoelectric effect is not the main factor causing changes in PL intensity. The PL measurements with positive remnant polarization with P (VDF-TrFE) are shown in Figure S2. Furthermore, several mechanisms are used to account for the PL change under an electric field. According to the previous reports, some possible factors for PL variation include thermal effect,$^{35}$ and intervally charge transfer.$^{57,58}$ Thermal effects can be ruled out because the opposite PL changes observed at reverse gate voltage cannot be explained by the temperature dependent PL
The applied negative gate voltage is removed after polarizing P (VDF-TrFE) with different gate voltages and the remnant polarization value is 7 μC/cm². The blue lines are only added as an eye guide.

Figure 4. (a) Calculated energy band structure of the bilayer WSe₂. The dashed arrows imply the possible radiation recombination pathways. (b) Electron transfer effect under strong electric field.
We conducted relativistic DFT calculation of the electronic structure of the bilayer WSe₂, as shown in figure 4 (a). According to the DFT calculation, the indirect band emission involves valence band holes at the K point which is nearly degenerate with the band at the Γ point and conduction band electrons at Λ point. The energy difference between the K and Λ point is about 40 meV. Due to large binding energy and small energy difference between the K and Λ point, electron transfer between different energy valleys is possible under the action of electric field, as displayed in figure 4 (b). According to the quasi-Boltzmann distribution law, the number of photocarriers at Λ point dominates. Since the quantum efficiency of the direct transition is much higher than the quantum efficiency of the indirect transition, the increase in the number of electrons in the K energy valley will inevitably lead to an increase in the entire fluorescence intensity, and vice versa. To support this argument, theoretical estimations are given as follows. When an external electric field is applied on the completely unpolarized ferroelectric material, the electronic polarization of the ferroelectric material between the electric dipole moment P and external electric field \( \xi \) is approximately expressed by

\[
P = -P_0 \tanh \left( \frac{\xi}{\xi_0} \right),
\]

where \( P_0 \) is the maximum polarization and \( \xi_0 \) denotes the hardness of the ferroelectric material. It should be noticed here the minus sign is determined by the setup of the apparatus in the experiment.

By solving the rate equation of the exciton populations of the direct and indirect exciton states (see supplementary material for further details), the population of the direct exciton state proportional to the PL peak intensity is given as

\[
n_e(\xi) \approx \beta \exp \left( - \frac{\delta + \Delta}{k_B T} \right) \frac{R(\xi)(1+r)}{R(\xi)r+1},
\]

where \( \delta \) is the energy difference of the conduction band extreme between K point and Λ point, \( \Delta \) is the energy gap of the bilayer WSe₂, \( k_B \) is Boltzmann constant and \( T \) is the temperature.

Here, \( r = \frac{\beta g_e}{g_s} \exp \left( \frac{\delta}{k_B T} \right) \) depends on the ratio between the excitation rates of the indirect exciton \( g_s \) and direct excitons \( g_e \), which is modified by a Boltzmann factor as well as the oscillating strength ratio \( \beta \) between the direct exciton and the indirect exciton. According to the transferred charge effect based on the effective electron temperature, the external field dependent ratio can be expressed as

\[
R(\xi) = \exp(-k \tanh \left[ \frac{\xi}{\xi_0} \right])
\]

with the dimensionless parameter
The electric dipole in the bilayer WSe$_2$ $p$ and the dielectric constant of the WSe$_2$ $\varepsilon$.

Figure 5 shows the integrated PL intensity of direct and indirect excitons as a function of the gate voltage. The calculated PL intensities are indicated by the red solid lines; the calculated intensities well reproduce the experimental results. The fitting parameters used here is $k = 1.24$, $\varepsilon_0 = 46.67$ eV, $\delta = 0.11$, $\Delta = 1.53$ eV, and $k_B T \approx 25.9$ meV for the room temperature $T = 300$ K. If the parameters in Eq. (4) are chosen as $p \approx 3 \times 10^8$ C·m and the internal electric field $P_0 / \varepsilon \approx 10^9$ V/m $^{27}$, the calculated dimensionless parameter $k \approx 1.19$ which is consistent with the fitting one.

Figure 5. The PL intensity of bilayer WSe$_2$ at different gate voltage. Circle represents PVDF top-gate data, star represents SiO$_2$ back-gate data. Solid lines show the calculated PL intensity curves calculated by solving the rate equations.

In addition, the direct exciton and indirect exciton emission intensity were extracted by Gaussian fitting method, shown. The fitting results show that the peak and FWHM of direct and indirect excitons are basically unchanged with the change of applied voltage (figure S5). As shown in figure 6, the PL intensity ratio of direct and indirect excitons decrease gradually with the applied bias change from negative 150 to positive 150. As the applied negative bias voltage goes up, the $\Lambda$ valley electrons will transfer to K valley via electric-assisted intervally scattering effect. As a
result, the carrier population at K point is increased and the direct exciton radiative transition is enhanced. This result is consistent with the intervallary carrier transfer model described above.

**Conclusion**

In summary, we observe that PL intensity in bilayer WSe$_2$ substantially reduced in position gate voltage and increased in negative gate voltage with increasing the electric field strength. The observed PL change is attributed to the transfer of intervally electrons from conduction band extremum K to Λ due to small energy difference (~55 meV) between K and Λ, and readjust the recombination path. These results provide a method for continuous improving the optical performance of optoelectronic devices based on bilayer WSe$_2$, and create an opportunity for hybrid integration of ferroelectric materials and 2D TMDCs as optical switch.

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