nature portfolio

Peer Review File

Electrical switching of spin-polarized light-emitting diodes based on a 2D CrI_3/hBN/WSe_2 heterostructure

Corresponding Author: Dr Xiao-Xiao Zhang

Reviewer comments:

Reviewer #1

(Remarks to the Author)
In this manuscript, the authors introduce an atomically thin spin-LED device. This device is composed of a heterostructure consisting of a monolayer of WSe2 and a few-layer antiferromagnetic CrI3, separated by a thin hBN tunneling barrier. Due to the spin-filtering effect induced by the CrI3/hBN layers, carriers passing through the barrier become spin-polarized, with their spin orientations closely tied to the magnet states of CrI3. These spin-polarized carriers are then directed into specific valleys within WSe2, as governed by the valley-dependent optical selection rule, resulting in the emission of circularly polarized light. Additionally, the authors demonstrate the device's noteworthy electrical tuning capabilities, including the ability to reverse electroluminescent circular polarization through the application of an electrostatic field. This tunability stems from the electrical adjustability of the few-layer CrI3 magnetization. Overall, the present work shows its novelty, and all experiments are meticulously designed. I recommend its publication in Nature Communications after addressing the following comments.
1. Based on the top gate-dependent EL data (Fig. S2), the authors observe a quenching of the EL signal when the WSe2 is tuned to a p-doped state. Consequently, they posit that the EL generation process can be elucidated through the band diagram presented in Fig. 1e. However, for additional validation of the correctness of the band diagrams depicted in Fig. 1e, it would be beneficial for the authors to conduct measurements of bottom gate-dependent current-voltage (I-V) transport alongside EL, followed by a comprehensive analysis of the obtained data.
2. In the PL spectra (Fig. 1f), multiple peaks are observed. I recommend that the authors delve into a discussion regarding the physical origin of these emission states. Additionally, Figure 1f presents a comparison between PL and EL spectra. However, it remains unclear whether the observed EL primarily stems from excitons, trions, or other contributors.
3. The authors report a maximum EL polarization of approximately 40% observed in the trilayer CrI3 device. In contrast, the circular polarization of neutral exciton PL is only around 23%, obtained under resonant optical excitation. However, it seems counterintuitive that the degree of EL polarization could exceed PL polarization. Additionally, the authors assert that the magnetic tunneling junctions could achieve near unity spin filter efficiency. However, the evidence or reasoning supporting this conclusion is not clearly articulated.
4. Compared to the EL polarization (Fig. 3c), the PL is oppositely polarized (Fig. 3d). The authors explain this opposite polarization is originated from the spin-dependent charge transfer, which quenches the valley/spin-polarized exciton with carriers' spin aligned with the CrI3 spin orientation. However, it is noteworthy that the presence of a hBN layer between WSe2 and CrI3 may hinder charge transfer, potentially enabling excitons to directly recombine and generate PL, distinct from the scenario described in reference 20. Furthermore, the degree of PL polarization in Figure 3d appears relatively small compared to existing literature. Additionally, it is important to consider that Figure 3d was conducted under a magnetic field of 2 Tesla, raising questions about the potential influence of magnetic control on valley pseudospin (Nature Physics 11, 148–152 (2015)). Therefore, further investigation is warranted before drawing conclusive statements regarding the observed PL polarization and its relation to charge transfer.
5. Drawing from Reference 24, it is suggested that a gate field has the potential to induce a transition from layered antiferromagnetic (AFM) to ferromagnetic (FM) states in bilayer CrI3. However, it appears that such a transition is not observable in Figure 4b-c, as evidenced by RMCD trace 2 and EL helicity not dropping to near zero. It would be beneficial for the authors to provide insights into the reasons behind this discrepancy.
6. The efficiency of spin-filtering could see a significant boost with increasing CrI3 layer thickness. It would be beneficial for the authors to establish the relationship between the degree of EL polarization and the thickness of CrI3 layers.

Reviewer #2

(Remarks to the Author)
The authors demonstrate spin-polarized light-emitting diodes based on heterostructure of monolayer WSe2 and few-layer

antiferromagnetic CrI3, separated by a thin hBN tunneling barrier. The electroluminescence exhibits a high degree of circular polarization that follows the CrI3 magnetic states. They also demonstrate an efficient electrical tuning, including a sign reversal, of the electroluminescent circular polarization by applying an electrostatic field. The results are interesting. The work has both scientific significance and application potential. The authors should make some revision according to the following questions/comments.
1. The authors claim that the WSe2 is n type, and CrI3 is p type. Please provide more experimental data, e.g. gate transfer curves of both WSe2 and CrI3 field-effect transistors, to confirm this. As far as I know, usually WSe2 behaves as p type and CrI3 behaves as n type. Besides, in Fig.1d, the I increases when a negative voltage is applied to the top gate (Vtg), also suggesting that the WSe2 be p type.
2. In Fig.1d, what the condition of the back gate, Vbg=0 or suspended?
3. In Fig.1e, what the conditions of the top and back gates? They are grounded or suspended?
4. To better under the tunneling current behavior of the device, the authors should provide the Ibias-Vbias curves at various Vbg.
5. There are some technical mistakes about subscript and blank space.

Reviewer #3

(Remarks to the Author)
The authors study a new spin LED structure consisting of monolayer WSe2 separated from few-layer CrI3 by a BN tunneling barrier. They demonstrate circularly polarized EL that is correlated with the CrI3 magnetization as demonstrated by magnetic field sweeps for devices with bilayer or trilayer CrI3. They demonstrate and explain how the EL has opposite circular polarization to the PL. Finally, they show how the EL can be tuned with gate voltage by relying on gate-dependent spin flip transitions at fixed finite magnetic fields. There are a few other works demonstrating circularly polarized EL using monolayer valley semiconductors, one with (Ga,Mn)As/WS2 and the other with FGT/BN/WSe2 (refs 7 and 8). Conceptually, these established works, in particular the FGT study, are quite similar to the present manuscript. Furthermore, the present work naturally extends previous works on CrI3/WSe2 structures that have shown an impact on the WSe2 PL polarization via CrI3 magnetization. Nevertheless, the work should be of significance to the field because of a few different advancements. The major achievement is the demonstration that the EL circular polarization can be tuned substantially with a gate voltage (under a fixed magnetic field). Usually, this requires other less technologically useful controls such as magnetic field or temperature changes. The EL helicity can also flip sign via gate voltage, although this process is not electrically reversible and so is not nearly as interesting. Another advancement is that the EL circular polarization can be quite large (~40% for trilayer CrI3 device), whereas typical spin LED devices usually only have single digit polarization percentages. Overall, the work is significant, the data support the claims, and the methodology is sound. Because of this, I can recommend publication after the authors address my minor comments below.

1. Do the authors understand why the degree of EL polarization is significantly higher for the trilayer sample(s)? It is not clear why there should be a such a difference between samples with bilayer vs trilayer spin injection when they are both in their saturated magnetization states.

2. The authors provide data for additional bilayer CrI3 devices, which show a max of 10-25% circular polarization depending on the device. This is important data to show the reproducibility of the phenomenon. Did the authors perform similar reproducibility experiments on the trilayer CrI3 device? It would also be worthwhile to show polarization-resolved EL spectra for the trilayer sample(s) in the SI similar to Fig. 2b.

3. Is the EL polarization electrically tunable in a reversible manner at the ~2T spin flip transitions in the trilayer CrI3 device?

Reviewer #1 (Remarks to the Author):

In this manuscript, the authors introduce an atomically thin spin-LED device. This device is composed of a heterostructure consisting of a monolayer of WSe2 and a few-layer antiferromagnetic CrI3, separated by a thin hBN tunneling barrier. Due to the spin-filtering effect induced by the CrI3/hBN layers, carriers passing through the barrier become spin-polarized, with their spin orientations closely tied to the magnet states of CrI3. These spin-polarized carriers are then directed into specific valleys within WSe2, as governed by the valley-dependent optical selection rule, resulting in the emission of circularly polarized light. Additionally, the authors demonstrate the device's noteworthy electrical tuning capabilities, including the ability to reverse electroluminescent circular polarization through the application of an electrostatic field. This tunability stems from the electrical adjustability of the few-layer CrI3 magnetization. Overall, the present work shows its novelty, and all experiments are meticulously designed. I recommend its publication in Nature Communications after addressing the following comments.

Reply: We really appreciate the referee's positive assessment of the novelty of this work and your recommendation to publish! We have provided more analysis and experimental data in the revised manuscript's main text and Supplementary Information according to the suggestions, which greatly improves the manuscript's quality.

1. Based on the top gate-dependent EL data (Fig. S2), the authors observe a quenching of the EL signal when the WSe2 is tuned to a p-doped state. Consequently, they posit that the EL generation process can be elucidated through the band diagram presented in Fig. 1e. However, for additional validation of the correctness of the band diagrams depicted in Fig. 1e, it would be beneficial for the authors to conduct measurements of bottom gate-dependent current-voltage (I-V) transport alongside EL, followed by a comprehensive analysis of the obtained data.

Reply: We thank the referee for the helpful advice. Following the suggestion, we measured the back gate-dependent I_{bias}-V_{bias} curves in one of the bilayer CrI_3 devices (while keeping the top gate fixed at 0 V), as shown in Fig. R1, which shows the tunneling current had minimal dependence within the range of the applied back gate voltages. This is expected and can be understood considering the large density of states (flat bands) of the CrI_3 band edges. With the finite electrostatic doping tuning capability, the corresponding Fermi level shifts in CrI_3 is much smaller than that can be achieved in WSe_2 and, therefore, do not have a significant impact on the I-V. We also measured the EL emission under different back gate voltages (Fig. R1c) following the suggestion, which does not have significant variation because of the constant tunneling current at different back gate voltages (Fig. R1b). If a larger doping modulation can be achieved in CrI_3 with, e.g., chemical doping, the heterostructure band alignment can be further verified, which will be of interest for future experiments.

In the revised submission, we include the back gate dependence of I-V and EL in the Supplementary Information (Fig. S2), and added the corresponding discussion in the main text.

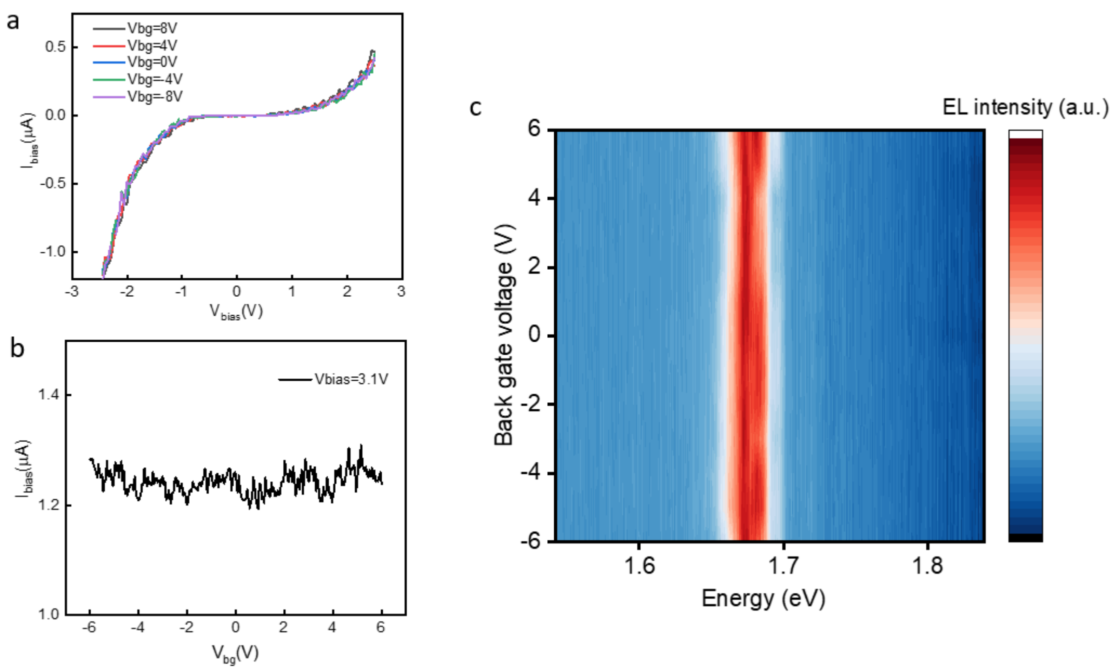

Figure R1. (a) I_V characteristics at different back gate voltages when $V_{tg} = 0V$. (b) Back gate dependent I_{bias} at a fixed $V_{bias} = 3.1V$, $V_{tg} = 0V$. (c) Back gate dependent EL when $V_{bias} = 3.1V$, $V_{tg} = 0V$. The slight fluctuations in intensity come from the fluctuations of I_{bias}.

2. In the PL spectra (Fig. 1f), multiple peaks are observed. I recommend that the authors delve into a discussion regarding the physical origin of these emission states. Additionally, Figure 1f presents a comparison between PL and EL spectra. However, it remains unclear whether the observed EL primarily stems from excitons, trions, or other contributors.

Reply: We fully agree we should present further analysis of the EL exciton contributions. In Fig. R2(a), which is also the same PL spectrum we showed in Fig. 1f (taken atV_{tg} = 0V, slightly n-doped), we first marked the exciton assignment according to their different energy splitting compared to the neutral bright exciton X_0, using the previously reported results in like https://www.nature.com/articles/s41467-020-14472-0 and https://www.nature.com/articles/s41467-018-05917-8 . The WSe_2 crystal quality in these devices is lower because the bulk WSe_2 was not flux-grown and contained more defects. Combined with the unintentional strain during the heterostructure transfer, the PL spectrum showed broad linewidths and merged localized emission peaks that are not present in "clean" samples.

To compare the EL with the PL spectrum, we chose the EL spectrum that contains multiple peaks and plotted it in a log scale to enhance small features, as shown in Fig. R2(b). Different devices show slightly different EL spectra, but the strongest emission features are around 1.65eV. There is no visible signal at the neutral bright exciton energy range. There are very small peaks near 1.7 eV that may be assigned to bright trion states in Fig. R2(b). However, we also note that the presence of tunneling current during the EL measurements leads to enhanced carrier screening, which can give rise to overall redshifts of exciton peaks and reductions of exciton binding energies (compared to PL measurements). The differences in screening conditions make it difficult to assign exciton peaks accurately in this EL device. Given the applied WSe_2 doping, the EL emission largely comes from charged exciton species. As for the relevance of valley polarization of the various excitonic states that contribute to the EL, we note that the injected *hole* carrier here are spin-polarized, which can give rise to excitonic polarization (both bright and dark) of the same valley/circular polarization.

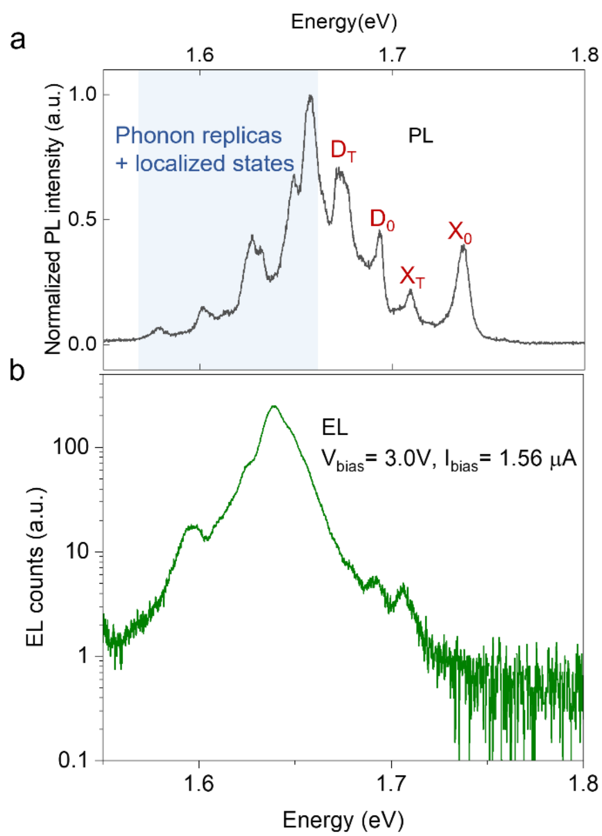

Figure R2. (a) Assignment of exciton species in PL spectrum and (b) Comparison with EL spectrum.

We added Fig. S3 (containing Fig. R2) in the revised submission to address the exciton assignment issue.

3. The authors report a maximum EL polarization of approximately 40% observed in the

trilayer CrI3 device. In contrast, the circular polarization of neutral exciton PL is only around 23%, obtained under resonant optical excitation. However, it seems counterintuitive that the degree of EL polarization could exceed PL polarization. Additionally, the authors assert that the magnetic tunneling junctions could achieve near unity spin filter efficiency. However, the evidence or reasoning supporting this conclusion is not clearly articulated.

Reply: We thank the referee for pointing out potential confusion in the EL & PL circular polarization comparison. We fully agree that it's not sufficient to just compare with the neutral exciton PL (of ~23% polarization). In the revised manuscript, we include the measurements of PL circular polarization (CP) at different doping levels with a near-resonant 633nm cw laser. As shown in Fig.R3(c), the bright n-type trion has a CP of ~ 40%, close to the highest CP observed in EL devices. Fig. R3(a) provides an example of circularly polarized PL measured at V_{tg} = 1V, with Fig. R3(b) showing the corresponding CP as a function of the photon wavelength. We also note that if the laser energy is closer to the exciton resonance (~710 nm), a slightly higher CP can be obtained.

We estimated that the spin filtering efficiency (across hBN/CrI_3) in some of our EL devices is close to 100% based on the comparison of maximum PL and EL circular polarization. In the PL excitation and emission process, 100% circular polarized on-resonant optical excitation gives rise to only < 50% polarized emission mainly because of the depolarization through the rapid intervalley exchange process (PhysRevB.89.205303 (2014), Nature Phys 12, 677–682 (2016)). A similar valley/spin depolarization within the WSe_2 will occur for the excitonic EL process. The obtained ~ 40% circular polarization in EL indicates the injected carriers into the WSe_2, and therefore, the electrically generated spin polarization is comparable to the ~ 100% valley- and spin-polarized optical excitation. This is a rough estimate because it ignores the possible depolarization differences under different screening environments when comparing the EL and PL processes.

We added a section in the revised Supplementary Information (Fig. S7) to include the gate-dependent PL polarization and provide a comparison between the PL and EL polarization. We have also added discussion in the main text to further compare the measured circular polarization in PL and EL.

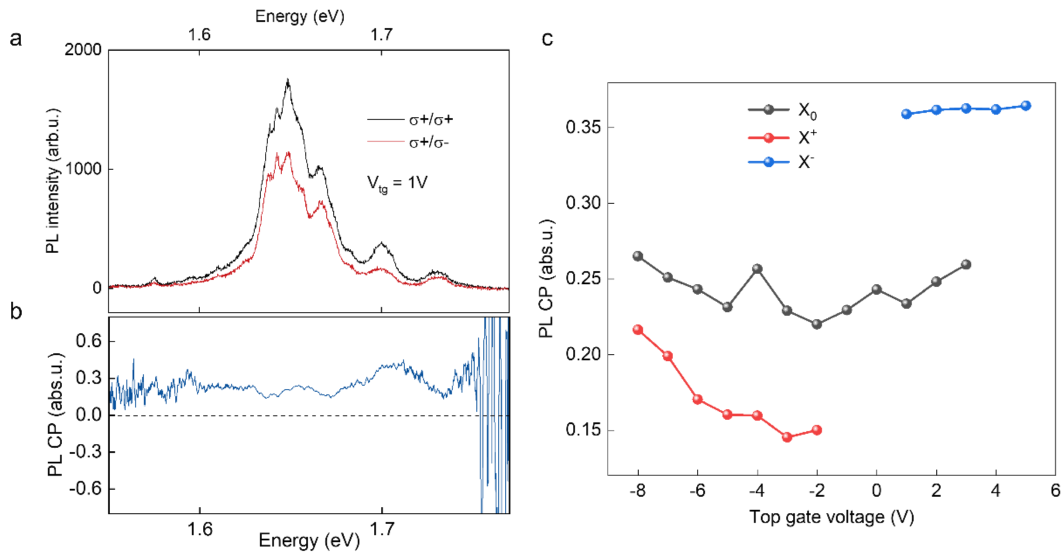

Figure R3. (a) Polarization-resolved PL spectra of WSe_2 at V_{tg} =1 and (b) the corresponding circular polarization (CP) extracted as a function of the photon energy. (c) Extracted CP for bright neutral and trion excitons as a function of the top gate voltages.

4. Compared to the EL polarization (Fig. 3c), the PL is oppositely polarized (Fig. 3d). The authors explain this opposite polarization is originated from the spin-dependent charge transfer, which quenches the valley/spin-polarized exciton with carriers' spin aligned with the CrI3 spin orientation. However, it is noteworthy that the presence of a hBN layer between WSe2 and CrI3 may hinder charge transfer, potentially enabling excitons to directly recombine and generate PL, distinct from the scenario described in reference 20. Furthermore, the degree of PL polarization in Figure 3d appears relatively small compared to existing literature. Additionally, it is important to consider that Figure 3d was conducted under a magnetic field of 2 Tesla, raising questions about the potential influence of magnetic control on valley pseudospin (Nature Physics 11, 148–152 (2015)). Therefore, further investigation is warranted before drawing conclusive statements regarding the observed PL polarization and its relation to charge transfer.

Reply: We fully agree that the presence of hBN may affect the charge transfer process (described in ref 20), and therefore, it should be more appropriate to compare the EL polarization direction with the PL polarization of the WSe_2/CrI_3 heterostructure. We thank the referee for correctly pointing out the issues when comparing with the PL signals from WSe_2/hBN/CrI_3.

We performed the PL polarization measurements on a WSe_2/CrI_3 device without an hBN barrier. We obtained a high PL polarization (Fig. R4d), consistent with previous work (ref 20). Compared with spin injection induced polarization in the EL (with hBN barrier), the polarization of PL from spin-dependent charge transfer shows the opposite polarization direction, as shown in Fig. R4c and R4d. In comparison, the presence of the thin hBN tunneling barrier indeed hinders charge transfer, which explains why the degree of PL polarization in previous Fig. 3d is relatively small. The opposite polarization directions we verified with the WSe_2/CrI_3 device support our explanation based on different carrier injection/relaxation as depicted in Fig. 3e&f.

In the revised manuscript, we have added more analysis and replaced the previous Fig. 3d with a clear PL polarization (Fig. R4b) caused by charge transfer. We also made changes to the corresponding discussion in the main text.

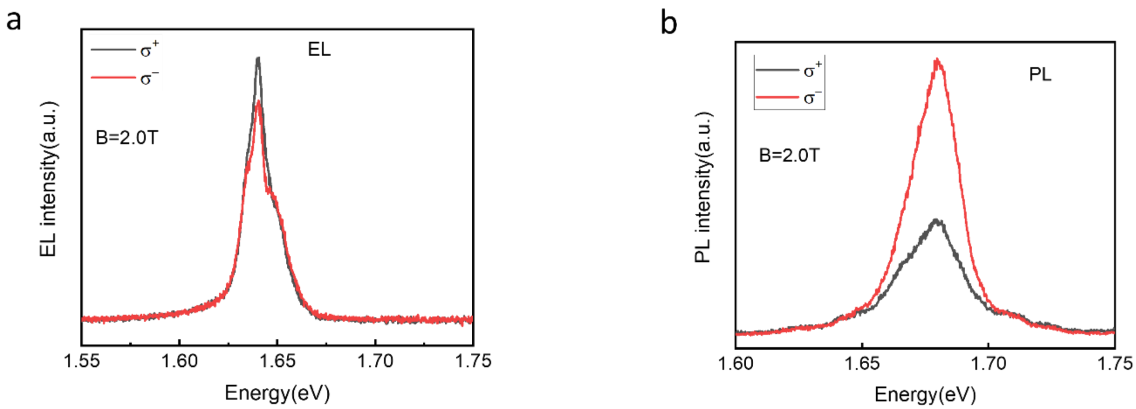

Figure R4. (a) Polarization of EL on a device with an hBN barrier. (b) Polarization of PL measured in heterostructure region on a device without hBN barrier. EL and PL exhibit opposite circular polarization.

5. Drawing from Reference 24, it is suggested that a gate field has the potential to induce a transition from layered antiferromagnetic (AFM) to ferromagnetic (FM) states in bilayer CrI3. However, it appears that such a transition is not observable in Figure 4b-c, as evidenced by RMCD trace 2 and EL helicity not dropping to near zero. It would be beneficial for the authors to provide insights into the reasons behind this discrepancy.

Reply: We thank the referee for pointing out this potential confusion. Based on Fig. 4c of Reference 22 and Fig. 2c of Reference 24, we can note that under a fixed magnetic field, a complete switching between AFM and FM states is induced only when the applied gate voltages are large enough (Reference 22: -30V to 30V with hBN as an insulating dielectric, and Reference 24: -50V to 50V with SiO_2 as the insulating dielectric of the bottom gate).

In our EL devices measurement, we applied a back gate voltage from -8V to 8V for our measurements to avoid destroying the device by a large gate voltage. Under the fixed magnetic field, both the RMCD and EL helicity show repeatable switching behaviors, though the switching is incomplete due to the limited gate voltage. As shown in Fig. R5, when we fixed the magnetic field at the point where the spin-flip transition starts, we can observe that the RMCD signal drops to near zero (trace 4). This further demonstrates that only a sufficiently large gate voltage can cause a complete switching. However, within our certain applied back gate voltage range, we can still clearly see efficient electrical tuning of RMCD and EL helicity. A complete and repeatable switching can be realized in future experiments after we demonstrate the electrical tunability in this prototypical device study.

We have added discussions in the revised manuscript's main text to address the partial switching.

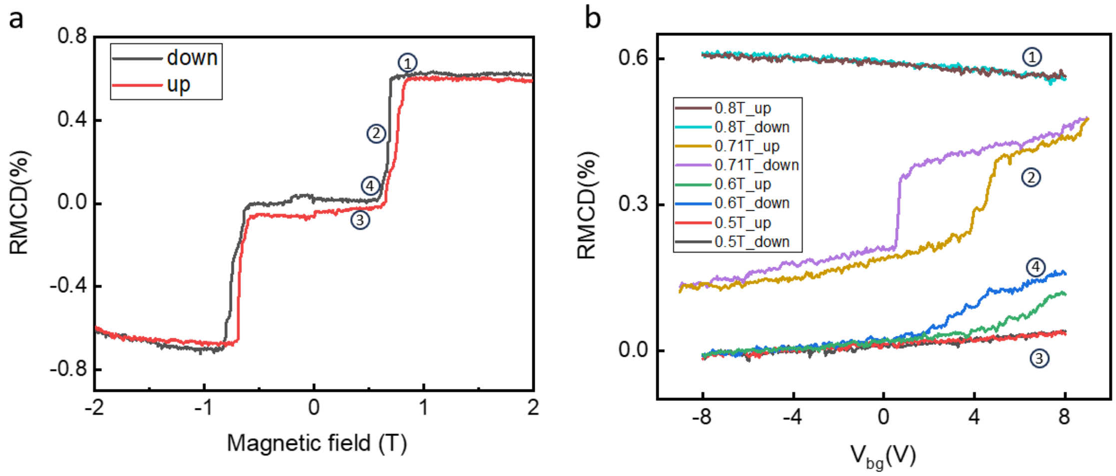

Figure R5. (a) Magnetic field dependent RMCD of bilayer CrI_3. (b) Back gate voltage control of RMCD at different fixed magnetic fields.

6. The efficiency of spin-filtering could see a significant boost with increasing CrI3 layer thickness. It would be beneficial for the authors to establish the relationship between the degree of EL polarization and the thickness of CrI3 layers.

Reply: Thanks for the reviewer's suggestion. We noticed that the saturation polarization for EL shows variation among different devices. After comparing multiple 2L and 3L devices, the maximum EL polarization does not seem to have a clear layer dependence, and the differences may lie in device and interface quality. Please see the summary of maximum EL polarization in Table 1. We have measured four bilayer CrI_3 devices and three trilayer CrI_3 devices (For one of the trilayer CrI_3 devices (D7), the EL helicity above the second spin-flip transition was not measured because the device broke down). From these data, it is difficult to establish a relationship between EL polarization and the thickness of CrI_3 layer, because the saturation polarization of one trilayer CrI_3 device (D6) is smaller than that of two bilayer CrI_3 devices (D2 and D4). In comparison, another trilayer device (D5) has a larger saturation polarization than the bilayer devices.

Future studies to establish the optimization conditions for better interfaces and device qualities will be very beneficial towards practical application but is beyond the current scope of this work.

We added Table 1 in the revised Supplementary Information and comments in the main text to discuss the variation in EL polarization in different devices.

Bilayer Devices	Saturation EL polarization (after SF)	
D1	10%	
D2	26%	
D3	10%	
D4	20%	
Trilayer Devices	**After first SF**	**After second SF**
D5	20%	40%
D6	8%	18%
D7	6%	NA

Table 1. Saturation polarization among different devices

Reviewer #2 (Remarks to the Author):

The authors demonstrate spin-polarized light-emitting diodes based on heterostructure of monolayer WSe2 and few-layer antiferromagnetic CrI3, separated by a thin hBN tunneling barrier. The electroluminescence exhibits a high degree of circular polarization that follows the CrI3 magnetic states. They also demonstrate an efficient electrical tuning, including a sign reversal, of the electroluminescent circular polarization by applying an electrostatic field. The results are interesting. The work has both scientific significance and application potential. The authors should make some revision according to the following questions/comments.

Reply: We appreciate the reviewer's positive comments. We have provided more discussions and experimental data in the revised manuscript and Supplementary Information. The following points are addressed below:

1. The authors claim that the WSe2 is n type, and CrI3 is p type. Please provide more experimental data, e.g. gate transfer curves of both WSe2 and CrI3 field-effect transistors, to confirm this. As far as I know, usually WSe2 behaves as p type and CrI3 behaves as n type. Besides, in Fig.1d, the I increases when a negative voltage is applied to the top gate (Vtg), also suggesting that the WSe2 be p type.

Reply: Thank you for your suggestions. The WSe_2 doping in our devices is controlled through the top gate while the WSe_2 is connected to the ground electrodes. The WSe_2 layer doping can be tuned from neutral to p-doped or n-doped based on the applied voltages, which is also verified in the gate-dependent photoluminescence measurement. When the doping level changes inside the WSe_2, negative and positive trion signals will appear in PL, while the neutral exciton signal quenches. The optical detection of TMD doping level has been widely applied in various optical and optoelectronic studies since earlier work that identified the different trion states (e.g. https://www.nature.com/articles/nmat3505 and https://www.nature.com/articles/ncomms2498). In Fig.R6, we compare the top gate dependent PL of the monolayer WSe_2 region and the heterostructure region that are in the same device, with marked positions of the neutral exciton (X^0), positive trion (X^+), negative trion (X^-) and neutral dark exciton (D^0). For the monolayer region, the charge neutral point is located near 0V, while for the heterostructure region, the neutral point has shifted to near negative -2.5V due to interactions with the CrI_3 layer. Most of the EL measurements were conducted under V_{bias}=2.5V, V_{tg}=1V with WSe_2 grounded and zero back gate (V_{bg} = 0V). In this applied voltage range, the WSe_2 is n-doped based on the separately assigned trion PL emission, which can be seen in Fig. R6(b). We also want to clarify that we didn't assume the doping state of the CrI_3 layer. The type-II band alignment (when V_{bias} = 0V) between the WSe_2 and CrI_3, as illustrated in the upper panel of Fig. 1e was confirmed in previous studies of WSe_2/CrI_3 heterostructures like https://www.science.org/doi/10.1126/sciadv.1603113 and https://www.nature.com/articles/s41563-020-0713-9 . When generating EL signals, the injected carriers into the WSe_2 through from the CrI_3/hBN layer are hole carriers.

Electrical measurements of the doping levels in each of the WSe_2 and CrI_3 layers will be beneficial in obtaining a comprehensive understanding of the electrical bands across the heterostructure. However, it will require a more complicated electrode layout (6 drain and source electrodes for three regions: WSe_2, CrI_3 and WSe_2 over the heterostructures). In addition, the CrI_3 is known to have very low mobility, which makes gate transfer curve type of measurements difficult. We thus consider these electrical measurements to be outside the scope of our current report.

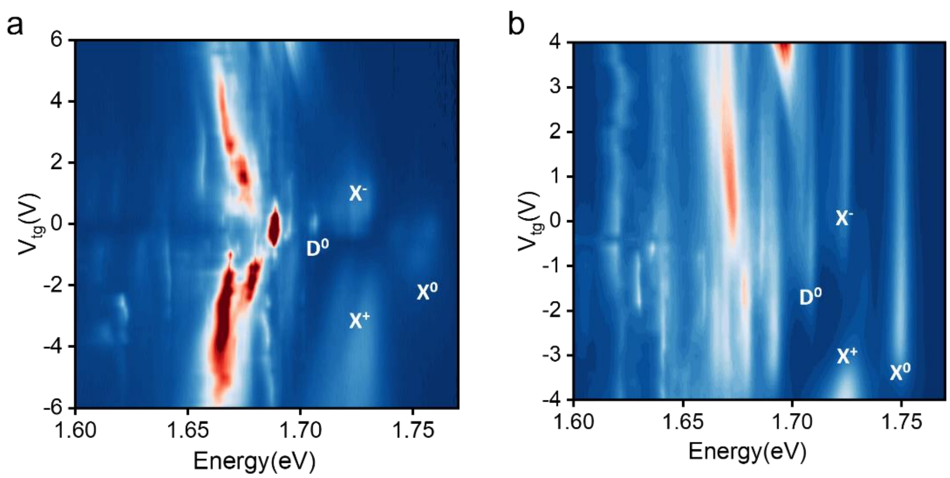

Figure R6. Top gate dependent PL spectra measured on the monolayer WSe_2(a) and heterostructure region(b).

We added Fig. S3 (a)&(b), which is the same as Fig. R6 in the Supplementary Information, to provide a detailed assignment of neutral and doped regimes based on the top gate dependent PL measurements.

2. In Fig.1d, what the condition of the back gate, Vbg=0 or suspended?

Reply: In Fig.1d measurement, the back gate V_{bg} is kept at zero.

We now emphasize this important information now both in the main text and in the figure captions.

3. In Fig.1e, what the conditions of the top and back gates? They are grounded or suspended?

Reply: Figure 1e is a schematic plot to illustrate the EL generation process. It depicts the corresponding scenario for EL generation with positive V_{bias} and the missing of the EL signal with negative V_{bias}. The back gate is always grounded, and the applied back voltages onto CrI_3 is not strong enough to affect the EL signals (see our detailed reply to Referee 1, Q1, and the next question). The top gate corresponds to the range of voltages that allow EL generation. As indicated by Fig. S2, it will correspond to the range of $V_{tg} > -2V$.

We add the specified experimental conditions corresponding to the Fig. 1e illustration caption in the revised manuscript.

4. To better under the tunneling current behavior of the device, the authors should provide the Ibias-Vbias curves at various Vbg.

Reply: Thanks for the reviewer's advice. We performed measurements of I_V curves at various V_{bg} and also measured the back gate dependent I_{bias} at a fixed V_{bias} on one of the devices, as shown in Fig. R7. The results depicted in Fig. R7 show that the back gates have almost no effect on the current. This is expected and can be understood considering the large density of states (flat bands) of the CrI_3 band edges. With the finite electrostatic doping tuning capability, the corresponding Fermi level shifts in CrI_3 are much smaller than that can be achieved in WSe_2 and, therefore, do not have a significant impact on the I-V. The corresponding EL as a function of back gate voltages can be found in Fig. R1c.

We add the I_V curves at different back gate voltages data and more analysis in the revised Supplementary Fig. S2.

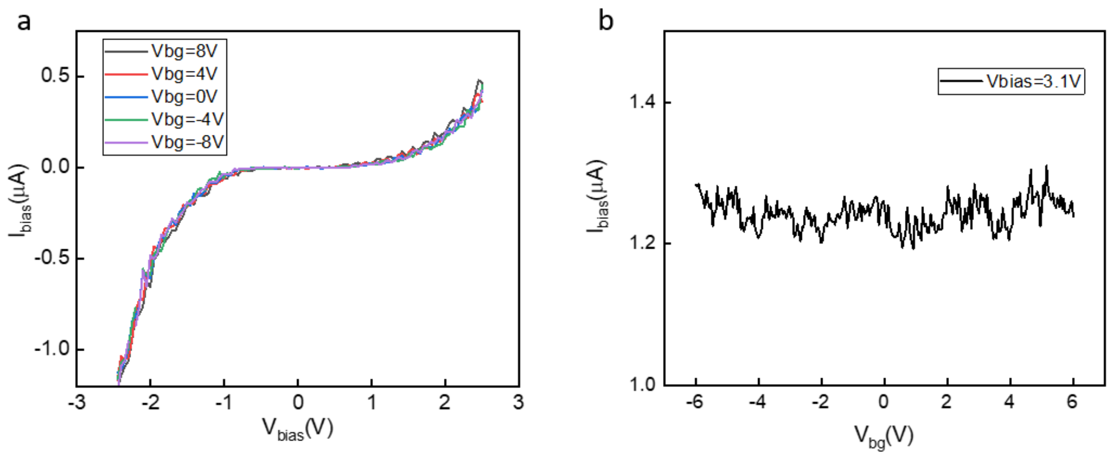

Figure R7. (a) I_V characteristics at different back gate voltages. (b) Back gate dependent I_{bias} at a fixed V_{bias}.

5. There are some technical mistakes about subscript and blank space.

Reply: We thank the referee for the careful and thorough reading. We have corrected them in the revised manuscript.

Reviewer #3 (Remarks to the Author):

The authors study a new spin LED structure consisting of monolayer WSe2 separated from few-layer CrI3 by a BN tunneling barrier. They demonstrate circularly polarized EL that is correlated with the CrI3 magnetization as demonstrated by magnetic field sweeps for devices with bilayer or trilayer CrI3. They demonstrate and explain how the EL has opposite circular polarization to the PL. Finally, they show how the EL can be tuned with gate voltage by relying on gate-dependent spin flip transitions at fixed finite magnetic fields. There are a few other works demonstrating circularly polarized EL using monolayer valley semiconductors, one with (Ga,Mn)As/WS2 and the other with FGT/BN/WSe2 (refs 7 and 8). Conceptually, these established works, in particular the FGT study, are quite similar to the present manuscript. Furthermore, the present work naturally extends previous works on CrI3/WSe2 structures that have shown an impact on the WSe2 PL polarization via CrI3 magnetization. Nevertheless, the work should be of significance to the field because of a few different advancements. The major achievement is the demonstration that the EL circular polarization can be tuned substantially with a gate voltage (under a fixed magnetic field). Usually, this requires other less technologically useful controls such as magnetic field or temperature changes. The EL helicity can also flip sign via gate voltage, although this process is not electrically reversible and so is not nearly as interesting. Another advancement is that the EL circular polarization can be quite large (~40% for trilayer CrI3 device), whereas typical spin LED devices usually only have single digit polarization percentages. Overall, the work is significant, the data support the claims, and the methodology is sound. Because of this, I can recommend publication after the authors address my minor comments below.

Reply: We are very encouraged by and appreciate the positive assessment of our work. We have provided more analysis and experimental data in the revised manuscript and Supplementary Information. The following points are addressed below:

1. Do the authors understand why the degree of EL polarization is significantly higher for the trilayer sample(s)? It is not clear why there should be a such a difference between samples with bilayer vs trilayer spin injection when they are both in their saturated magnetization states.

Reply: Thanks for the reviewer's comment. We noticed that the saturation polarization for EL shows variation among different devices. After comparing multiple 2L and 3L devices, the maximum EL polarization does not seem to have a clear layer dependence and the differences may lie in device and interface quality. Please see the summary of maximum EL polarization in Table 1. We have measured four bilayer CrI_3 devices and three trilayer CrI_3 devices (For one of the trilayer CrI_3 devices (D7), the EL helicity above the second spin-flip transition was not measured because the device broke down). From these data, it is difficult to establish a relationship between EL polarization and the thickness of CrI_3 layer, because the saturation polarization of one trilayer CrI_3 device (D6) is smaller than that of two bilayer CrI_3 devices (D2 and D4). In comparison, another trilayer device (D5) has a larger saturation polarization than the bilayer devices.

Future studies to establish the optimization conditions for better interfaces and device qualities will be very beneficial towards practical application but is beyond the current scope of this work.

We added Table 1 in the revised Supplementary Information and comments in the main text to discuss the variation in EL polarization in different devices.

Bilayer Devices	Saturation EL polarization (after SF)	
D1	10%	
D2	26%	
D3	10%	
D4	20%	
Trilayer Devices	**After first SF**	**After second SF**
D5	20%	40%
D6	8%	18%
D7	6%	NA

Table 1. Saturation polarization among different devices

2. The authors provide data for additional bilayer CrI3 devices, which show a max of 10-25% circular polarization depending on the device. This is important data to show the reproducibility of the phenomenon. Did the authors perform similar reproducibility experiments on the trilayer CrI3 device? It would also be worthwhile to show polarization-resolved EL spectra for the trilayer sample(s) in the SI similar to Fig. 2b.

Reply: We thank the referee for raising this important point. Yes, we also performed magnetic field-dependent RMCD and EL helicity measurements for another trilayer CrI_3 device, as depicted in Fig. R8a and b. Additionally, we present the corresponding polarization-resolved EL spectra, displayed in Fig. R8c and d.

We have provided additional experimental data from another trilayer CrI_3 device in the revised Supplementary Fig. S4.

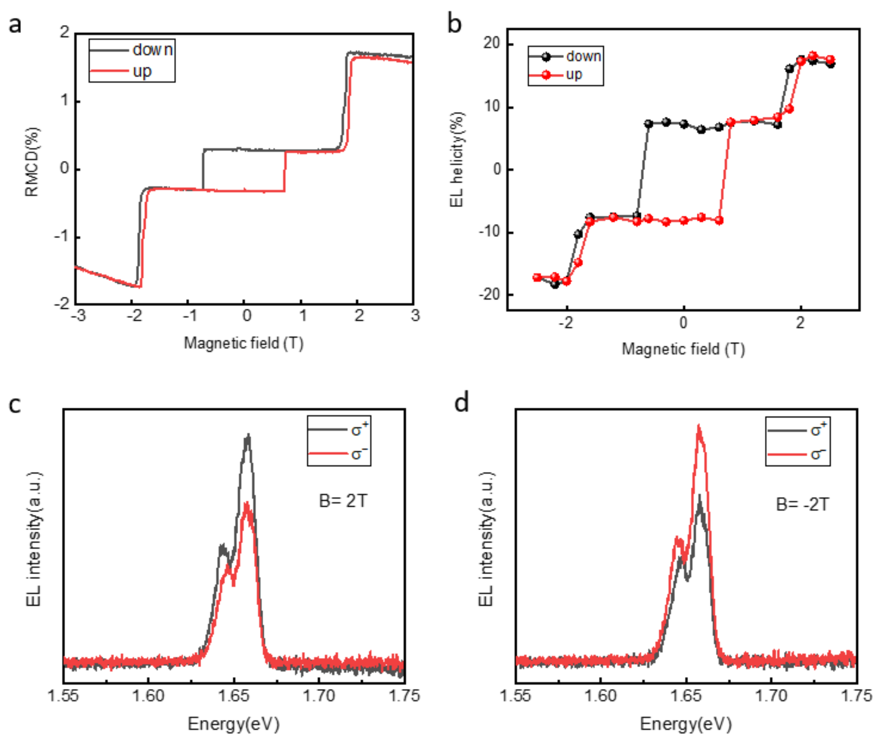

Figure R8. (a) RMCD signal and (b) EL helicity as a function of magnetic fields for trilayer CrI_3 device. (c) and (d) Polarization-resolved EL spectra under ±2T magnetic fields.

3. Is the EL polarization electrically tunable in a reversible manner at the ~2T spin flip transitions in the trilayer CrI3 device?

Reply: We thank the great suggestion to include more data. Yes, we measured the reversibility for the trilayer CrI_3 device near the spin-flip transition field (at 1.73T). As shown in Fig. R9, it also shows the repeatable switching of EL polarization with the back gate voltage as expected.

We have presented the experimental data in the revised Supplementary Fig. S8.

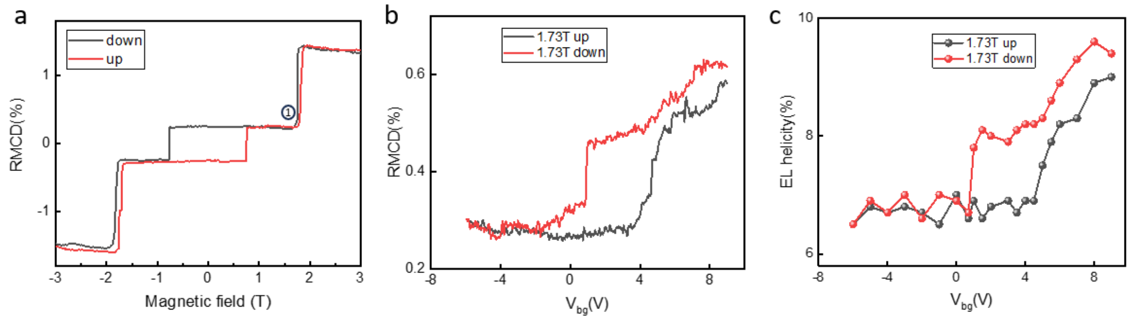

Figure R9. (a) Magnetic field dependent RMCD of trilayer CrI_3. (b) Back gate voltage control of RMCD of trilayer CrI_3 at a fixed magnetic field. (c) Corresponding EL helicity at different back gate voltages.

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors have adequately addressed my comments. Now, I would support its publication.

Reviewer #2

(Remarks to the Author)
I am satisfied with the authors' response, and suggest the paper be published as it is.

Reviewer #3

(Remarks to the Author)
The authors have satisfactorily addressed my comments and revised the manuscript appropriately. I believe it is suitable for publication in Nature Communications.