

Spin-polarized light-emitting diodes based on CrI₃ operating without external spin injection

Corresponding Author: Professor Chang-Hua Liu

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This manuscript experimentally demonstrates a spin-polarized electroluminescent diode based on monolayer and bilayer CrI₃. Compared with conventional GaAs-based emitters, this device enables direct control of the emission helicity through the magnetic state of CrI₃, without relying on external spin injectors. This approach avoids the complex heterostructure engineering typically required to integrate spin injectors and suppress spin decoherence, while achieving a degree of circular polarization (20%) that surpasses most traditional spin-polarized LEDs. Overall, the work provides a promising platform for future on-chip spin-optoelectronic devices. After the following issues are properly addressed, I recommend its publication in Nature Communications.

1. The schematic in Fig. 3(c) may be easily confused with a band-structure diagram. If possible, the authors are encouraged to include a corresponding schematic based on band theory or crystal-field theory to clarify the underlying physical mechanism.
2. As the injection current increases, the emission peak redshifts, while both the linewidth and intensity increase. The authors attribute the redshift to Joule heating, but no direct evidence is provided. Moreover, the physical origin of the increased linewidth and intensity is not sufficiently explained. Additional analysis or experimental support is needed.
3. The sign reversal in the EL DOCP loop occurs at around ± 0.1 T, which is lower than the coercive field (0.17 T) obtained from RMCD and magneto-PL measurements. Is there an additional physical mechanism responsible for this discrepancy? For example, could the applied bias modify the magnetic properties of CrI₃? The authors should discuss this point or provide supporting evidence.
4. The DOCP decreases with increasing current. The authors attribute this to the weakening of CrI₃ magnetic order due to Joule heating, supported by simulations showing that the material temperature rises from 30 K to 36 K. However, this temperature is still far below the Curie temperature (45 K), making this explanation less convincing. Carrier injection can change the band occupation of CrI₃ and may intrinsically affect its magnetism; the authors are encouraged to consider and discuss this possibility.
5. Numerous studies have shown that bilayer CrI₃ undergoes an antiferromagnetic-to-ferromagnetic phase transition under applied bias. Since the present work involves bilayer CrI₃, the authors should discuss this effect and its potential implications for their experimental observations.

Reviewer #2

(Remarks to the Author)

The manuscript reports electrically driven, magnetically switchable circularly polarized electroluminescence from monolayer CrI₃. This is, to my knowledge, the first demonstration of a spin-LED using monolayer CrI₃ as the intrinsic emitter without relying on additional spin-polarizing TMD layers, and the ability to generate EL from monolayer CrI₃ at all is itself a technically nontrivial and interesting result. The integration of CrI₃ with graphene/hBN tunneling contacts is executed cleanly, and the optical–magnetic coupling they observe is consistent with the expected magnetic order of CrI₃. These aspects make the work a meaningful addition to the growing effort of vdW-magnet–based optoelectronics.

At the same time, the manuscript appears to substantially overstate both the conceptual novelty and the performance significance of the device. While the authors emphasize achieving ~20% DOCP and frame this as addressing the “limited

DOCP" in prior spin-LEDs, the paper does not explain why monolayer CrI₃ should fundamentally overcome those limitations that affect other spin-LED architectures. No conceptual mechanism is articulated. As a result, the motivational narrative lacks the scientific grounding needed to justify the authors' claims about broader impact. Without a mechanistic understanding, the work reads primarily as a materials demonstration rather than a solution to the DOCP bottleneck the authors highlight. In its current form, I do not feel it meets the level of conceptual clarity and novelty required for publication in Nature Communications.

This issue is especially problematic because a directly relevant prior work, Dang et al., Nat Commun 15, 6799 (2024), already reports 26–40% DOCP in CrI₃/TMD spin-LEDs, substantially higher than the ~20% reported here. The absence of any discussion of this work creates a misleading impression that 20% DOCP represents a breakthrough in vdW spin-LEDs, when in fact the state of the art already exceeds it. Moreover, the authors repeatedly compare their results only to traditional III–V and II–VI spin-LEDs, omitting the most relevant vdW-magnet-based devices, including linearly polarized EL in NiPS₃ (Lebedev et al., Nat Commun 16, 10550 (2025)) and electrically controlled polarization in bilayer CrI₃/TMD devices. A fair, complete comparison is essential for evaluating the manuscript's contributions. I strongly encourage the authors to discuss their results holistically within the existing body of vdW-based spin-LEDs and clearly articulate what novelty remains. For example: does monolayer CrI₃ emission avoid the intervalley depolarization that limits the DOCP in CrI₃/TMD devices? If so, what is the physical mechanism?

The experimental support for the core claims is also limited. The key DOCP value is shown for only one monolayer device (and one bilayer device in the SI). Prior studies, including those the authors cite, have documented significant device-to-device variability. Without multiple devices or at least a clear demonstration of reproducibility, it is difficult to evaluate how representative the reported 20% polarization actually is. For a claim centered on "high DOCP," this lack of statistical confirmation is a notable weakness. Given that the previous investigation into CrI₃/TMD devices showed considerable variation in the DOCP of different devices (see Dang et al. Supplementary Table 1), it would also be beneficial for the authors to include additional data from other supporting monolayer or bilayer devices to establish that the reported DOCP ~ 20% is reproducible across multiple samples rather than a single exceptional case.

Finally, because EL from monolayer CrI₃ has not been previously established, a deeper discussion of the emission mechanism, possible defect-assisted recombination, and device stability under bias would significantly strengthen the manuscript. As written, these aspects are treated only briefly, leaving the interpretation of the EL behavior incomplete.

Reviewer #3

(Remarks to the Author)

I have read the manuscript "Spin-polarized light-emitting diodes based on CrI₃ operating without external spin injection" by C.-C. Lu, et al. This work reports circularly polarized electroluminescence from CrI₃ tunneling devices. The data is of high quality, and the interpretation straightforward. Although somewhat similar results have been observed in WSe₂ / CrI₃ heterostructures, this is the first work to my knowledge to report spin-LED operation from a single 2D magnetic layer. Therefore, in my opinion, it deserves to be published in Nature Communications. Nevertheless, I have a couple comments the authors may consider:

- 1) Do the author's see any effect on the coercive field due to the applied bias voltage? That is, can the device be switched electrically (by changing the bias) if the external field is close to the coercive field?
- 2) The authors associate the drop in degree of circular polarization (DOCP) at high bias to heating effects. Yet, the temperature induced phase transition of Ising-type ferromagnetic monolayer CrI₃ is rather sharp, while the high-bias drop in DOCP is gradual. Perhaps the author's can clarify this discrepancy?
- 3) In my opinion, the first two figures could probably be combined into one. These two do not really report anything new (the circularly polarized CrI₃ photoluminescence was reported long ago in 2018) compared to the main results of the paper. However, I recognize that this point is entirely one of personal taste, so I leave it up to the authors to decide.

Reviewer #4

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors have satisfactorily addressed all of my questions and concerns. I therefore recommend the manuscript for publication.

Reviewer #2

(Remarks to the Author)

The authors have substantially revised the manuscript in response to my previous report. In particular, they have moderated several of the claims regarding device performance and clarified that the primary novelty lies in demonstrating circularly polarized electroluminescence originating from the intrinsic magneto-exciton coupling of monolayer CrI_3 , rather than in achieving a record degree of circular polarization. The revised manuscript now more clearly positions the work as an injector-free spin-LED architecture, where circular polarization arises from the magnetic order of the emitting layer itself. This clarification addresses a central concern raised in my previous review report.

The additional discussion regarding current-induced spectral changes and the possible influence of carrier injection on the magnetic properties of CrI_3 is also helpful. While some of these interpretations remain somewhat speculative, such as the bias-induced modification of exchange interactions, the authors now acknowledge these limitations and present them more cautiously.

Overall, the manuscript presents a technically solid experimental demonstration of electrically driven circularly polarized emission from monolayer CrI_3 heterostructures with graphene tunneling contacts. Although the achieved DOCP (~20%) is not exceptional compared to other vdW spin-LED platforms, the injector-free architecture represents an interesting design concept for spin-optoelectronic devices based on 2D magnets.

I therefore recommend publication after possible, but not mandatory, minor revisions. In particular, the authors may consider further clarifying the microscopic mechanism linking magnetic order to EL helicity, and discussing whether the recombination process under electrical injection is expected to preserve the same polarization behavior observed in photoluminescence.

Reviewer #3

(Remarks to the Author)

In my opinion, the authors suitably addressed my concerns. In particular, I appreciate that they made five additional monolayer CrI_3 devices - these are challenging devices to make and I commend their effort.

Reviewer #4

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

made.

In cases where reviewers are anonymous, credit should be given to 'Anonymous Referee' and the source.

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Point-by-point Responses to Reviewers' Comments

Reviewers' comments are in red; our responses are in blue.

Reviewer #1 (Remarks to the Author):

1. This manuscript experimentally demonstrates a spin-polarized electroluminescent diode based on monolayer and bilayer CrI₃. Compared with conventional GaAs-based emitters, this device enables direct control of the emission helicity through the magnetic state of CrI₃, without relying on external spin injectors. This approach avoids the complex heterostructure engineering typically required to integrate spin injectors and suppress spin decoherence, while achieving a degree of circular polarization (20%) that surpasses most traditional spin-polarized LEDs. Overall, the work provides a promising platform for future on-chip spin-optoelectronic devices. After the following issues are properly addressed, I recommend its publication in Nature Communications.

We thank the reviewer for the positive assessment of the paper and careful review of our manuscript. We have addressed all the comments below.

2. The schematic in Fig. 3(c) may be easily confused with a band-structure diagram. If possible, the authors are encouraged to include a corresponding schematic based on band theory or crystal-field theory to clarify the underlying physical mechanism.

We appreciate the reviewer's comment. In the revised manuscript, Fig. 3(c) has been updated to illustrate that the electroluminescence originates from transitions between the t_{2g} and e_g orbitals of Cr^{3+} . The revised figure caption also explicitly states that, due to crystal-field interactions, the d orbitals of Cr^{3+} split into three lower-energy t_{2g} orbitals (d_{xy} , d_{yz} , d_{xz}) and two higher-energy e_g orbitals ($d_{x^2-y^2}$, d_{z^2}). This clarification helps avoid confusion with a conventional band-structure diagram.

3. As the injection current increases, the emission peak redshifts, while both the linewidth and intensity increase. The authors attribute the redshift to Joule heating, but no direct evidence is provided. Moreover, the physical origin of the increased linewidth and intensity is not sufficiently explained. Additional analysis or experimental support is needed.

We thank the reviewer for this insightful and substantive comment. We agree that attributing the observed spectral evolution solely to Joule heating is not sufficient. In the revised manuscript, we have included additional experiments and analyses to clarify the physical origins of the observed

electroluminescence redshift, linewidth broadening, and intensity enhancement under electrical injection.

Specifically, as the injection current increases up to 50 μA , the EL peak exhibits a redshift from 1.14 to 1.17 μm accompanied by clear linewidth broadening (Fig. 3d–f). Although our finite-element simulations suggest that this current range increases the device temperature from 30 K to approximately 36 K (Supplementary Section 3), temperature-dependent photoluminescence measurements on monolayer CrI_3 indicate that such a temperature rise produce negligible peak redshift and linewidth broadening (revised Supplementary Section 4). Thus, as the reviewer suggested, Joule heating alone cannot fully explain the current-induced spectral changes in electroluminescence, and additional mechanisms must contribute. Notably, recent studies show that excitons in CrI_3 are highly localized, with emission described by Frenkel excitons, exciton polarons, or self-trapped excitons. While these models differ in mechanism, all predict that increasing exciton population can lead to linewidth broadening and peak redshift, via enhanced phonon sidebands in Frenkel excitons or increased lattice distortions in exciton polarons and self-trapped excitons. These exciton-related effects could thus contribute to the current-induced spectral changes in addition to heating.

Regarding the current-dependent emission intensity, we observe that the EL intensity increases sublinearly with the injection current (Fig. 3f). From a device perspective, this sublinear behavior is commonly seen in tunneling light-emitting diode structures, where a strong vertical electric field at high bias enhances direct tunneling of carriers between the graphene electrodes (Ref. 31-33). Consequently, the fraction of carriers undergoing radiative recombination decreases, reducing the contribution to light emission at high injection levels. From the perspective of the emitting material, CrI_3 , exciton–exciton annihilation under strong injection may further suppress the emission intensity, as recently reported (J. Chem. Phys. 161, 114706 (2024)), contributing to the observed sublinear scaling. These analyses and the associated references have been incorporated into the revised manuscript.

4. The sign reversal in the EL DOCP loop occurs at around ± 0.1 T, which is lower than the coercive field (0.17 T) obtained from RMCD and magneto-PL measurements. Is there an additional physical mechanism responsible for this discrepancy? For example, could the applied bias modify the magnetic properties of CrI_3 ? The authors should discuss this point or provide supporting evidence.

The sign reversal in the EL DOCP loop occurs at around ± 0.1 T, which is lower than the coercive field (0.17 T) obtained from RMCD and magneto-PL measurements. In original manuscript, we attribute this difference as RMCD and PL are sensitive to local magnetization at a focused spot, whereas EL integrates emission across the entire CrI_3 layer, effectively averaging over possible domain variations. We agree with the reviewer, however, that the applied bias can also play an important role. Specifically, increasing the bias voltage not only enhances the carrier injection rate but also raises the chemical potential of the graphene electrodes, enabling

carriers to populate higher-energy orbitals in CrI_3 , including the Cr d_{z^2} and $d_{x^2-y^2}$ orbitals as well as the I p_x and p_y orbitals. As suggested by recent literature (2D Mater. 12, 045025 (2025)), occupation of these orbitals can introduce antiferromagnetic exchange interactions, which weaken the overall ferromagnetic order in CrI_3 .

Notably, this bias-induced weakening of ferromagnetic order provides an additional explanation for why the EL DOCP sign reversal occurs at a lower magnetic field than that measured by RMCD and magneto-PL. To further support this interpretation, we measured EL DOCP loops at different bias voltages (added in Supplementary Section 7) and observed that the EL DOCP reverses sign at even lower magnetic fields as the bias voltage increases. The corresponding experimental results, discussion, and references have been incorporated into the revised manuscript.

5. The DOCP decreases with increasing current. The authors attribute this to the weakening of CrI_3 magnetic order due to Joule heating, supported by simulations showing that the material temperature rises from 30 K to 36 K. However, this temperature is still far below the Curie temperature (45 K), making this explanation less convincing. Carrier injection can change the band occupation of CrI_3 and may intrinsically affect its magnetism; the authors are encouraged to consider and discuss this possibility.

We agree with the reviewer that raising the injection current to 50 μA increases the device temperature from 30 K to ~ 36 K, slightly weakening the ferromagnetic order. However, this is still below the Curie temperature of monolayer CrI_3 (~ 45 K), so the material remains ferromagnetic. Thus, the rapid DOCP drop from 20% at 0.5 μA to 2.5% at 50 μA cannot be explained by Joule heating alone.

We further agree that carrier injection under high bias can intrinsically modify the magnetic properties of CrI_3 . As discussed in our response to point #4 and supported by recent theoretical work (2D Mater. 12, 045025 (2025)), the elevated chemical potential of the graphene electrodes enables carriers to populate higher-energy Cr d and I p orbitals. This altered band occupation enhances antiferromagnetic exchange interactions, thereby weakening the ferromagnetic order and contributing to the observed decrease in DOCP with increasing current (Fig. 4d). The corresponding discussion has been added to the revised manuscript.

6. Numerous studies have shown that bilayer CrI_3 undergoes an antiferromagnetic-to-ferromagnetic phase transition under applied bias. Since the present work involves bilayer CrI_3 , the authors should discuss this effect and its potential implications for their experimental observations.

We agree with the reviewer that numerous studies have reported an antiferromagnetic-to-

ferromagnetic phase transition in bilayer CrI₃ induced by electrostatic gating that tunes the doping level in CrI₃. In the literature, a common approach employs graphene/hBN/CrI₃/hBN/graphene heterostructures, in which the two graphene layers serve as top and bottom gates to electrostatically tune the doping in CrI₃. Although this device architecture is similar to that used in our spin-polarized emitters, the hBN layers in our device are only two layers thick and are introduced to enable efficient carrier tunneling rather than to function as gate dielectrics. Consequently, the graphene electrodes in our device do not provide effective electrostatic gating of CrI₃, and the structure cannot be regarded as a capacitive gating geometry. An alternative route for electrostatic gating could, in principle, be realized via the silicon back gate. In our device, however, the gate electric field is strongly screened by the graphene bottom electrode as well as by the large density of carriers injected into CrI₃ under bias. As a result, sweeping the silicon back-gate voltage does not produce observable changes in the EL DOCP hysteresis loop.

We note, however, that replacing SiO₂ with a high- κ dielectric could enhance the gating efficiency and potentially enable the antiferromagnetic-to-ferromagnetic transition, thereby providing a viable route for actively tuning the DOCP in spin-polarized light-emitting diodes. This perspective has been included in the revised conclusion section.

Additional Note: During the manuscript revision, we noticed that the y-axis in Fig. 3a (I–V characteristics) was mislabelled. An applied bias of ± 3 V corresponds to a current injection of ± 50 μ A, rather than ± 5 μ A as shown in the original manuscript. This has been corrected in the revised version, and we apologize for any confusion this may have caused.

Reviewer #2 (Remarks to the Author):

7. The manuscript reports electrically driven, magnetically switchable circularly polarized electroluminescence from monolayer CrI₃. This is, to my knowledge, the first demonstration of a spin-LED using monolayer CrI₃ as the intrinsic emitter without relying on additional spin-polarizing TMD layers, and the ability to generate EL from monolayer CrI₃ at all is itself a technically nontrivial and interesting result. The integration of CrI₃ with graphene/hBN tunneling contacts is executed cleanly, and the optical–magnetic coupling they observe is consistent with the expected magnetic order of CrI₃. These aspects make the work a meaningful addition to the growing effort of vdW-magnet–based optoelectronics.

We thank the reviewer for the positive assessment of the paper and careful review of our manuscript.

8. At the same time, the manuscript appears to substantially overstate both the conceptual

novelty and the performance significance of the device. While the authors emphasize achieving ~20% DOCP and frame this as addressing the “limited DOCP” in prior spin-LEDs, the paper does not explain why monolayer CrI₃ should fundamentally overcome those limitations that affect other spin-LED architectures. No conceptual mechanism is articulated. As a result, the motivational narrative lacks the scientific grounding needed to justify the authors’ claims about broader impact. Without a mechanistic understanding, the work reads primarily as a materials demonstration rather than a solution to the DOCP bottleneck the authors highlight. In its current form, I do not feel it meets the level of conceptual clarity and novelty required for publication in Nature Communications.

We thank the reviewer for this important comment. We agree that the achieved DOCP of ~20% is not, by itself, exceptionally high compared with conventional III–V/II–VI spin-LEDs, and that some van der Waals spin-LEDs have reported DOCP values approaching ~40%. Accordingly, we have removed the statement claiming that our work addresses the “limited DOCP” in prior spin-LED architectures.

We clarify that the novelty of our device does not lie in the absolute magnitude of the DOCP, but in the physical mechanism by which circularly polarized electroluminescence is generated. In our device, the DOCP originates from the intrinsic properties of the emitting layer, monolayer CrI₃. Specifically, the strong coupling between excitons and the magnetic order in CrI₃ leads to spin-dependent splitting of the ground-state excitons (Adv. Mater. 35, e2209513 (2023)). This intrinsic magneto-excitonic coupling directly produces circular polarization in the emitted light. As a result, circularly polarized electroluminescence is achieved without the use of an external spin injector, which is typically required in conventional spin-LEDs and imposes stringent demands on spin-polarized carrier injection and spin coherence preservation during transport.

We further emphasize that the reported ~20% EL DOCP should be regarded as an initial demonstration of this mechanism. Its potential improvement is discussed in detail in the point-to-point response to Comment #9, and the characteristics of excitonic emission and the underlying mechanisms in CrI₃-based devices are further elaborated in the point-to-point response to Comment #11.

9. This issue is especially problematic because a directly relevant prior work, Dang et al., Nat Commun 15, 6799 (2024), already reports 26–40% DOCP in CrI₃/TMD spin-LEDs, substantially higher than the ~20% reported here. The absence of any discussion of this work creates a misleading impression that 20% DOCP represents a breakthrough in vdW spin-LEDs, when in fact the state of the art already exceeds it. Moreover, the authors repeatedly compare their results only to traditional III–V and II–VI spin-LEDs, omitting the most relevant vdW-magnet-based devices, including linearly polarized EL in NiPS₃ (Lebedev et al., Nat Commun 16, 10550 (2025)) and electrically controlled polarization in bilayer CrI₃/TMD devices. A fair, complete comparison is essential for evaluating the manuscript’s contributions. I strongly encourage the authors to discuss their results holistically within the existing body of vdW-based spin-LEDs and

clearly articulate what novelty remains. For example: does monolayer CrI_3 emission avoid the intervalley depolarization that limits the DOCP in CrI_3 /TMD devices? If so, what is the physical mechanism?

We appreciate the reviewer's comment and agree that a fair and comprehensive comparison with prior van der Waals spin-LEDs is essential. In the revised Introduction, we have expanded the discussion of earlier studies using two-dimensional magnetic materials as spin injectors to generate spin-polarized carrier injection into TMDs and realize fully vdW spin-LED architectures. This can be realized by using Fe_3GeTe_2 as a spin source, yielding degree of circular polarization (DOCP) $\sim 8\%$ at 78 K, with nonvolatile DOCP sign reversal under a low magnetic field (~ 0.1 T). Alternatively, trilayer CrI_3 can function as a spin-filter barrier, producing EL from TMDs with a DOCP of up to $\sim 40\%$, achieved at 4 K under a 2 T magnetic field, which ensures CrI_3 converts from an antiferromagnetic to a ferromagnetic state and enables efficient spin filtering (Dang et al., Nat. Commun. 15, 6799 (2024)).

In addition, our revision emphasizes that prior vdW-magnet-based and conventional III–V/II–VI spin-LEDs rely on an external spin injector, which requires efficient spin-polarized carrier injection, preservation of spin coherence during transport, and spin-conserving radiative recombination in the emitter (a process affected by intervalley depolarization in TMDs). To overcome these limitations, an attractive strategy is to employ 2D magnets directly as the light-emitting medium, exploiting the intrinsic coupling between excitons and the magnetic order to produce polarized emission. We explicit mention that this concept has recently been validated in NiPS_3 , where electroluminescence exhibits an ultranarrow linewidth (~ 1 meV) and a high degree of linear polarization, arising from excitons coupled to the underlying antiferromagnetic order (Lebedev et al., Nat. Commun. 16, 10550 (2025)). We then highlight that, building on this paradigm, our work uses CrI_3 itself as the emitter, leveraging exciton–magnetization coupling to produce circularly polarized electroluminescence without the need for an external spin injector. We believe this revision clarifies the distinctions and highlights the novelty of our work within the context of vdW-based spin-LEDs.

In addition to revising the Introduction, we have updated the Conclusion to discuss potential strategies for increasing DOCP in our device. One approach is lowering the device temperature, which enhances CrI_3 magnetization and reduces thermally populated carriers between exciton states split by the magnetic order (Adv. Mater. 35, e2209513 (2023)), thereby increasing DOCP. Another strategy is to increase CrI_3 magnetization through p-type doping, which has been demonstrated experimentally and theoretically (2D Mater. 12, 045025 (2025) and Nat. Nanotech. 13, 549–553 (2018)).

Finally, we emphasize that although Dang et al. (Nat. Commun. 15, 6799 (2024)) report EL DOCP values up to $\sim 40\%$, there are several key differences with our work. First, the $\sim 20\%$ PL and EL DOCP reported here were measured at 30 K—higher than the 4 K used in Dang et al.—

and are limited by our current setup. In fact, prior study shows CrI_3 can reach PL DOCP above 50% at 15 K (Nat. Phys. 14, 277–281 (2018)), with even higher values expected at lower temperatures. Second, the device-to-device variation of EL DOCP in our work is relatively small compared with previous reports, reflecting that the DOCP in our devices is solely determined by magneto-excitonic coupling rather than by the more complex spin injection, transport, or recombination processes. Third, the ~40% DOCP reported in Dang et al. was obtained under an ~2 T magnetic field. The high field is required to convert the layered antiferromagnetic CrI_3 into a ferromagnetic state for efficient spin filtering. In contrast, in our devices, EL DOCP can be switched with only 0.1 T, and due to the non-volatile nature of the switching, ~20% DOCP can be sustained even in the absence of an applied magnetic field, facilitating practical device operation.

10. The experimental support for the core claims is also limited. The key DOCP value is shown for only one monolayer device (and one bilayer device in the SI). Prior studies, including those the authors cite, have documented significant device-to-device variability. Without multiple devices or at least a clear demonstration of reproducibility, it is difficult to evaluate how representative the reported 20% polarization actually is. For a claim centered on “high DOCP,” this lack of statistical confirmation is a notable weakness. Given that the previous investigation into CrI_3 /TMD devices showed considerable variation in the DOCP of different devices (see Dang et al. Supplementary Table 1), it would also be beneficial for the authors to include additional data from other supporting monolayer or bilayer devices to establish that the reported DOCP ~ 20% is reproducible across multiple samples rather than a single exceptional case.

We agree with the reviewer that demonstrating device-to-device reproducibility is important for assessing the representativeness of the reported DOCP. To address this concern, we fabricated and characterized five independent monolayer CrI_3 devices, all measured under identical operating conditions at 30 K. All devices exhibit clear circularly polarized electroluminescence, with DOCP values consistently ranging from ~18% to ~23%, indicating good device-to-device reproducibility rather than an isolated exceptional case. The corresponding data and discussion have been added to the revised manuscript and the Supplementary Section 6.

11. Finally, because EL from monolayer CrI_3 has not been previously established, a deeper discussion of the emission mechanism, possible defect-assisted recombination, and device stability under bias would significantly strengthen the manuscript. As written, these aspects are treated only briefly, leaving the interpretation of the EL behavior incomplete.

We appreciate the reviewer’s comment and agree that a more in-depth discussion of the emission mechanism is necessary. In the revised manuscript, we have updated Fig. 3c to schematically illustrate that the emission originates from intra-atomic transitions between the t_{2g} and e_g orbitals of Cr^{3+} . The revised figure caption explicitly states that, due to crystal-field interactions,

the Cr^{3+} d orbitals split into three lower-energy t_{2g} orbitals (d_{xy} , d_{yz} , d_{xz}) and two higher-energy e_g orbitals ($d_{x^2-y^2}$, d_{z^2}).

In the revised main text, we further discuss that these intra-atomic d – d transitions are highly localized, and their emission may exhibit characteristics associated with exciton polarons, Frenkel excitons, or self-trapped excitons, as suggested by recent studies (Adv. Mater. 35, e2209513 (2023), Nano Lett. 22, 8755–8762 (2022) and Phys. Rev. Lett. 133, 046903 (2024)). While these models differ in mechanism, all predict that increasing exciton population can lead to linewidth broadening and peak redshift, via enhanced phonon sidebands in Frenkel excitons or increased lattice distortions in exciton polarons and self-trapped excitons. These exciton-related effects could thus contribute to the current-induced EL spectral changes (Fig. 3e–f) in addition to heating. The corresponding discussion and references have been included in the revised manuscript.

Beyond the spectral evolution, we also expand the discussion of the current dependence of the EL intensity (Fig. 3f, lower panel). In the original manuscript, this behavior was attributed to enhanced carrier tunneling between the graphene electrodes under strong vertical electric fields at high bias, which reduces the fraction of injected carriers contributing to radiative recombination. In the revised manuscript, we further note that exciton–exciton annihilation in the emitting CrI_3 layer may also occur under intense carrier injection, as suggested by recent studies (J. Chem. Phys. 161, 114706 (2024)), providing an additional nonradiative decay channel. Taken together, these effects can account for the observed sublinear scaling of the EL intensity with injection current.

Extending this discussion, we also discuss how increasing carrier injection can affect the magnetization of CrI_3 and thus EL DOCP (Fig. 4d) in the revised manuscript. Specifically, we elaborate that increasing the bias voltage not only enhances the carrier injection rate, which can lead to heating effects, but also raises the chemical potential of the graphene electrodes, allowing higher-energy states in CrI_3 to be populated by injected carriers, including the $\text{Cr } d_{z^2}$ and $d_{x^2-y^2}$ orbitals as well as the $\text{I } p_x$ and p_y orbitals. As suggested by recent theoretical work (2D Mater. 12, 045025 (2025)), occupation of these states can introduce antiferromagnetic exchange interactions that weaken the overall ferromagnetic order of CrI_3 . Given that the exciton splitting in CrI_3 is closely tied to its magnetization, these effects provide a plausible explanation for the observed decrease in EL DOCP with increasing injection current (Fig. 4d).

Regarding device stability, we include Supplementary Section 6 to show that both EL intensity and DOCP remain stable after 48 hours of continuous operation. While longer-term evaluation will be important for practical device applications, these results demonstrate the preliminary robustness of our devices.

Additional Note: During the manuscript revision, we noticed that the y-axis in Fig. 3a (I–V characteristics) was mislabelled. An applied bias of ± 3 V corresponds to a current injection of ± 50 μ A, rather than ± 5 μ A as shown in the original manuscript. This has been corrected in the revised version, and we apologize for any confusion this may have caused.

Reviewer #3 (Remarks to the Author):

12. I have read the manuscript "Spin-polarized light-emitting diodes based on CrI₃ operating without external spin injection" by C.-C. Lu, et al. This work reports circularly polarized electroluminescence from CrI₃ tunneling devices. The data is of high quality, and the interpretation straightforward. Although somewhat similar results have been observed in WSe₂ / CrI₃ heterostructures, this is the first work to my knowledge to report spin-LED operation from a single 2D magnetic layer. Therefore, in my opinion, it deserves to be published in Nature Communications. Nevertheless, I have a couple comments the authors may consider:

We thank the reviewer for the positive assessment of the paper and careful review of our manuscript.

13. Do the author's see any effect on the coercive field due to the applied bias voltage? That is, can the device be switched electrically (by changing the bias) if the external field is close to the coercive field?

We thank the reviewer for this insightful question. By measuring the EL DOCP hysteresis loops under different applied bias voltages, we indeed observe that the EL DOCP switches sign at a lower magnetic field as the bias voltage on the graphene electrodes increases (added in Supplementary Section 7). This behavior indicates that increasing the applied bias reduces the coercive field of CrI₃. Consequently, when the external magnetic field is tuned close to the coercive field, modulation of the applied bias can indeed switch the sign of the EL DOCP. However, higher bias also increases the EL intensity while reducing the amplitude of the DOCP, which may limit the practical utility of bias-controlled DOCP switching.

In addition to this experimental demonstration, we discuss the physical origin in the revised manuscript. Specifically, the bias-induced reduction in coercivity can be attributed to the elevated chemical potential of the graphene electrodes under high bias, which allows injected carriers to populate higher-energy orbitals in CrI₃, including the Cr dz^2 and dx^2-y^2 orbitals as well as the I p_x and p_y orbitals. As suggested by recent theoretical studies (2D Mater. 12, 045025 (2025)), occupation of these states can introduce antiferromagnetic exchange interactions, weakening the overall ferromagnetic order. This weakening provides a physical basis for the bias-dependent reduction in coercivity and also links to the observed EL DOCP reduction at higher bias (relevant to point-to-point response #14).

In addition to varying the bias across the graphene electrodes, we also investigated EL DOCP hysteresis loops under different silicon back-gate voltages but did not observe any discernible changes. This lack of gate dependence can be attributed to strong electrostatic screening by the bottom graphene electrode and the high carrier density injected from the graphene electrodes, which limits the effectiveness of back-gate modulation of coercivity.

14. The authors associate the drop in degree of circular polarization (DOCP) at high bias to heating effects. Yet, the temperature induced phase transition of Ising-type ferromagnetic monolayer CrI₃ is rather sharp, while the high-bias drop in DOCP is gradual. Perhaps the author's can clarify this discrepancy?

We thank the reviewer for pointing this out. Indeed, as the reviewer notes, the measured DOCP gradually decreases from 20% to 2.5% as the injection current increases up to 50 μ A (Fig. 4d). This gradual behaviour is inconsistent with the typical temperature dependence of CrI₃ magnetization, which shows a sharp drop near the Curie temperature ($\sim$ 45 K). It is also notable that our finite-element simulations indicate the applied current raises the device temperature only from 30 K to $\sim$ 36 K, well below the Curie temperature, suggesting that Joule heating alone cannot fully account for the observed reduction in DOCP.

In the revised manuscript, we attribute this discrepancy to an additional mechanism closely related to the bias-induced effects described in response #13. Specifically, the elevated chemical potential of the graphene electrodes under high bias allows carriers to populate higher-energy Cr d and I p orbitals, enhancing antiferromagnetic exchange interactions. This effect weakens the overall ferromagnetic order in CrI₃. Together with modest current-induced heating, these effects plausibly explain the gradual decrease in EL DOCP with increasing injection current. The revised manuscript now incorporates this explanation along with supporting references.

15. In my opinion, the first two figures could probably be combined into one. These two do not really report anything new (the circularly polarized CrI₃ photoluminescence was reported long ago in 2018) compared to the main results of the paper. However, I recognize that this point is entirely one of personal taste, so I leave it up to the authors to decide.

We thank the reviewer for this suggestion. We agree that Fig. 2 presents results similar to those already reported in the literature. However, combining it with Fig. 1 into a single figure would require reducing the figure size to fit the journal's format, which could compromise clarity and readability. For this reason, we have kept them as separate figures.

Additional Note: During the manuscript revision, we noticed that the y-axis in Fig. 3a (I–V characteristics) was mislabelled. An applied bias of $\pm$ 3 V corresponds to a current injection of $\pm$ 50 μ A, rather than $\pm$ 5 μ A as shown in the original manuscript. This has been corrected in the revised version, and we apologize for any confusion this may have caused.

Reviewer #4 (Remarks to the Author):

16. I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

We thank the reviewer for co-reviewing this manuscript and for the valuable input.