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Peer Review File

Optimal spin-charge interconversion in graphene through spin-pseudospin entanglement control

Corresponding Author: Mr Joaquin Medina Dueñas

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

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
This manuscript examine the effect of tunable spin-orbit coupling on spin-charge inter-connection. This result numerically evaluate the strength of the effect which could be useful for potential application. The calculations are correct and sufficient to justify the result. I think the result is of interest of the particular field though. Also, despite the current work is novel, I believe in general the effect of spin-orbit coupling on spin-charge interchange has been explored.

Reviewer #2

(Remarks to the Author)
In this manuscript J.M. Duenas and others put forward an interesting idea that optimized spin-charge interconversion (SCI) in monolayer graphene-based structures with Rashba-Dirac electrons can be achieved by promoting the role of the Kane-Mele interaction (KM). It was previously recognized that a single Fermi surface of Rashba-Dirac electrons is favorable for uncompensated rise of the Rashba Edelstein effect (REE), but an upper limit was set due to the spin-pseudospin entanglement-induced suppression of the Rashba spin textures [Phys. Rev. Lett. 119, 569 196801 (2017), Ref.36]. In the current work, it is noted that when the KM term is accounted for, the so-called pseudospin gap appears in the spectrum, when the single Fermi surface features spin helix structure free of spin-pseudospin entanglement. As a result, this allows one to further improve the intrinsic effectiveness of REE and SCI conversion, as well as to get finite SHE. The discussed results are corroborated (i) using the Bastin-Kubo calculations (ii) performing advanced numerical simulations of a graphene monolayer with finite Anderson disorder based on real-space tight-binding modelling and the Kernel Polynomial method. Like I mentioned in the beginning, I find the work interesting and useful for a community, its conclusions are well justified and the manuscript is well written.

I have some minor comments to be addressed before the manuscript is fully suitable for publication

1. Throughout the paper, the Rashba constant is taken to be 10 meV; the KM term is varied and also achieves 10 meV. Could these magnitudes be justified more rigorously? So far, 10 meV for both terms seems fairly above what is obtained experimentally and from first-principles.

2. Also, typical Fermi energies for a monolayer graphene in transport experiments are somewhat larger than 5-10 meV, lying in tens and hundreds of meV range. What is the corresponding concentration of electrons for the Fermi energy to remain within 10 meV gap?
At such a low doping (corresponding to 5-10 meV energy range) I would expect the thermal distribution of both electrons and holes to be very important. Can this affect the obtained conclusions about the intrinsic effectiveness of SCI and REE? What are the upmost temperatures when the intrinsic effectiveness of SCI remains optimal and not thermally distorted?

3. For the introduction section and the discussion of the spin-pseudospin correlation effects, I believe another important example could be mentioned, which is the coupled spin-pseudospin dynamics which results in the electric field induced spin resonance and spin-flip absorption from [Phys. Rev. B, 109, L201406 (2024)].

4. In lines 249-254, I would suggest to explain better what is the actual outcome of the simulations. Why would Sy and Jx remain unaffected by disorder within the pseudogap? I would naively expect both quantities to scale linearly with the total relaxation time [transport time] that should decrease at stronger disorder, i.e. 1/tau ~ n |W|^2 [n is the 2D density of Anderson impurities]. Sure, the ratio of REE is to stay mostly unaffected, but the disorder is still to affect Sy and Jx magnitudes.

5. Strictly speaking, Eq.5 is not actually a clean limit: 5a is nothing but the Drude current which diverges at vanishing scattering (i.e. in the true clean limit). My understanding is that Eq.4 and 5 based analysis corresponds to a simplified treatment of disorder: The scattering time is simply defined by the broadening of the single-particle Green's function poles rather than by self-consistent evaluation from the vertex equations. This, however, does not transform the diffusive transport regime from Eq.5b, so saying "beyond the clean limit" in line 231 [and other places] might be misleading.

6. Is it true that the definition of the REE efficiency [line 188] has an "intrinsic" character: It reflects the intrinsic degree of the spin conversion from the Drude current to the nonequilibrium spin polarization? Does this "intrinsic" quantity actually determine the "external" effectiveness of REE and some related effects [e.g. spin-orbit torques in a neighboring magnet]? Would the usage of the drift velocity in line 188 [instead of the Fermi velocity] better reflect the actual amount of spin polarization we can generate and use in real devices?

Reviewer #3

(Remarks to the Author)
Please see the attached PDF

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 Communications Physics 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 #2

(Remarks to the Author)
The Authors have done a substantial work answering my questions. I find the corrections of the manuscript very satisfactory and can now recommend the revised version for the publication.

Reviewer #3

(Remarks to the Author)
See attached file

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 Communications Physics initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Response to the Reviewer's remarks to authors, for manuscript COMMSPHYS-25-1672

We here respond to the reviewer's remarks to the authors. We first respond to the reviewer's questions and comments point-by-point, indicating the modifications in the manuscript resulting from these discussions. We finally summarise all changes performed to the revised manuscript.

We use the following convention:

"italic and quoted": reviewer's remarks

regular: author's response

We also highlight in **bold writing** the modifications to the manuscript resulting from the Reviewer's comments.

Alongside this response, we provide the revised manuscript where all modifications are clearly indicated.

REVIEWER #1: point-by-point response

> *"This manuscript examines the effect of tunable spin-orbit coupling on spin-charge inter-connection. This result numerically evaluate the strength of the effect which could be useful for potential application. The calculations are correct and sufficient to justify the result. I think the result is of interest of the particular field though. Also, despite the current work is novel, I believe in general the effect of spin-orbit coupling on spin-charge interchange has been explored."*

We thank the Reviewer for their comments. The Reviewer values the novelty of our work and the interest it will raise within the research field, and recognizes the validity of our method and results. Furthermore, the general topic of spin-orbit coupling effects on spin-charge interconversion in graphene has received widespread attention throughout the years. In this context, the Reviewer values our work as a novel step and deems it is of interest to the community.

REVIEWER #2: point-by-point response

> *"In this manuscript J.M. Duenas and others put forward an interesting idea that optimized spin-charge interconversion (SCI) in monolayer graphene-based*

structures with Rashba-Dirac electrons can be achieved by promoting the role of the Kane-Mele interaction (KM). It was previously recognized that a single Fermi surface of Rashba-Dirac electrons is favorable for uncompensated rise of the Rashba Edelstein effect (REE), but an upper limit was set due to the spin-pseudospin entanglement-induced suppression of the Rashba spin textures [Phys. Rev. Lett. 119, 569 196801 (2017), Ref.36]. In the current work, it is noted that when the KM term is accounted for, the so-called pseudospin gap appears in the spectrum, when the single Fermi surface features spin helix structure free of spin-pseudospin entanglement. As a result, this allows one to further improve the intrinsic effectiveness of REE and SCI conversion, as well as to get finite SHE. The discussed results are corroborated (i) using the Bastin-Kubo calculations (ii) performing advanced numerical simulations of a graphene monolayer with finite Anderson disorder based on real-space tight-binding modelling and the Kernel Polynomial method. Like I mentioned in the beginning, I find the work interesting and useful for a community, its conclusions are well justified and the manuscript is well written.

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"1. Throughout the paper, the Rashba constant is taken to be 10 meV; the KM term is varied and also achieves 10 meV. Could these magnitudes be justified more rigorously? So far, 10 meV for both terms seems fairly above what is obtained experimentally and from first-principles."

We thank the Reviewer for their observation, as it motivates us to better justify the system we study.

Proximity-induced Rashba SOC in graphene/TMD bilayers has been found to range from ~1 meV [Wang, et al. *Nature Communications* **6** 8339 (2015)] up to ~15 meV [Wang, et al. *Phys. Rev. X* **6** 041020 (2016); Wang, et al. *Nano Letters* **19** 10 7028-7038 (2019), Sun, et al. *Nature Communications* **14** 3771 (2023)]. On the other hand, Kane-Mele SOC is negligible in said heterostructures, but emerges in graphene/TI bilayers with a strength of a few meV [Khokhriakov, et al. *Science Advances* **4** eaat9349 (2018)], and can also be enhanced by decoration with adatoms [Weeks, et al. *Phys. Rev. X.* **1** 021001 (2011)] or nanoparticles [Hatsuda, et al. *Science Advances* **4** 11 (2018)], reaching the ~10 meV scale. Based on this evidence, the values explored for the SOC parameters in the manuscript lie within the experimental range. **In light**

of the Reviewer's comment, we have modified the paragraph including Eq. (1) (page 2), where the material platforms for realising the proposed system are discussed. The reviewed discussion contains all the references presented in this answer, and clearly indicates the orders of magnitude that have been found for the SOC parameters.

We additionally note that, even though we take the Rashba SOC parameter as 10 meV throughout the manuscript, the conclusions obtained hold for smaller SOC values. Indeed, the figures of merit studied in the manuscript, such as the Rashba-Edelstein effect efficiency and the spin Hall conductivity, are insensitive to a rescaling of the SOC parameters within the low-energy regime. As shown by the results presented in section S1 of the Supplementary Information, the spin and pseudospin textures are invariant to a rescaling of the SOC parameters with an additional rescaling of the energy by the same factor, accounting for the rescaling of the pseudogap. Therefore, the REE efficiency, which is composed of a ratio between two Fermi surface magnitudes, is insensitive to a rescaling of the SOC parameters, and so is the spin Hall conductivity which corresponds to an intrinsic Fermi sea magnitude.

We finally note that a platform presenting relevant Rashba and Kane-Mele SOC simultaneously has not been yet identified. In this sense, we remark our work constitutes a theoretical proposal, which can hopefully promote the search for graphene platforms beyond state of the art in order to observe this novel physics.

> *"2. Also, typical Fermi energies for a monolayer graphene in transport experiments are somewhat larger than 5-10 meV, lying in tens and hundreds of meV range. What is the corresponding concentration of electrons for the Fermi energy to remain within 10 meV gap?*
> *At such a low doping (corresponding to 5-10 meV energy range) I would expect the thermal distribution of both electrons and holes to be very important. Can this affect the obtained conclusions about the intrinsic effectiveness of SCI and REE? What are the upmost temperatures when the intrinsic effectiveness of SCI remains optimal and not thermally distorted?"*

The Reviewer is correct that the native Fermi level in such devices lies typically some tens of meV far from the Dirac point due to doping. Nevertheless, graphene's Fermi level can be largely tuned by applying a gate voltage. For instance, in a graphene/WS_2 spin-charge interconversion experiment [Benítez, et al. *Nature Materials* **19** 170-175

(2019)] the carrier density is modulated within the $\pm 10^{12}$ cm^{-2} range by applying a gate voltage, which corresponds to a ±100 meV modulation of the Fermi level [Foa Torres, et al, "Introduction to Graphene-Based Nanomaterials: From Electronic Structure to Quantum Transport", *Cambridge University Press* (2020)]. Therefore, the pseudogap regime is accessible by gating in such devices. In particular, the ±10 meV window about charge neutrality point corresponds to a $\pm 10^{10}$ cm^{-2} carrier concentration.

The Reviewer raises an interesting point regarding the effects of the carrier thermal distribution at finite temperature. We perform the transport calculations for the REE within the constant broadening approximation (called the clean-limit in the previous version of the manuscript, and corrected in the revised version, following the Reviewer's comment Nº5), considering a finite temperature which only affects the carrier thermal distribution. The corresponding longitudinal current J_x and non-equilibrium spin density S_y correspond to convolutions of the zero temperature result with the derivative of the Fermi-Dirac distribution, as indicated by Eq. (7a). For the transport velocity, we recall that it is defined based on the Einstein relation, and therefore the density of states appearing therein (see Methods section) must also be convoluted by the Fermi-Dirac distribution derivative in order to reflect the amount of carriers available for transport [Di Ventra, "*Electrical Transport in Nanoscale Systems*", Cambridge University Press (2010)]. The results for the REE efficiency are shown in Fig. R1-(a, b) below. We observe that the REE efficiency in the optimal $\lambda_{KM}=\lambda_R$ case maintains its peak value up to T ~ 30 K, whereas for the $\lambda_{KM}=0$ case the peak REE efficiency is reduced for any finite temperature. We conclude that the system is in general sensitive to thermal effects due to the small size of the Rashba pseudogap, where the proposed spin-charge-interconversion phenomena are most relevant. Nevertheless, in the optimal case we propose, the plateau-shaped maximum grants further robustness to the REE against the effects of finite temperature. We also compute the spin Hall conductivity in this case, shown in Fig. R1-(c, d), where we observe a similar behaviour. For $\lambda_{KM}=\lambda_R$, the SHE conductivity is finite and presents the same sign throughout the entire pseudogap, which grants a further robustness against the thermal spreading of the electron occupation. **We have included the present analysis in the supplementary information of the revised manuscript.**

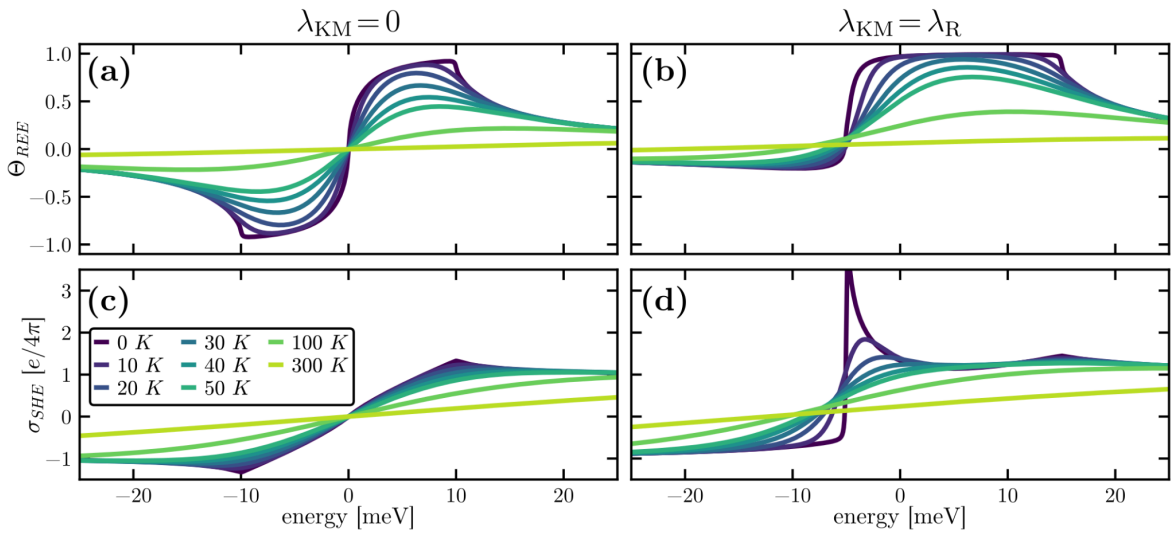

Fig R1: REE efficiency (a, b) and SHE conductivity (c, d) at finite temperature for λ_{KM}=0 (a, c) and λ_{KM}=λ_R (b, d).

> *"3. For the introduction section and the discussion of the spin-pseudospin correlation effects, I believe another important example could be mentioned, which is the coupled spin-pseudospin dynamics which results in the electric field induced spin resonance and spin-flip absorption from [Phys. Rev. B, 109, L201406 (2024)]."*

We thank the Reviewer for their suggestion. **We have included the mentioned phenomenon and its reference in the revised manuscript.**

> *"4. In lines 249-254, I would suggest to explain better what is the actual outcome of the simulations. Why would Sy and Jx remain unaffected by disorder within the pseudogap? I would naively expect both quantities to scale linearly with the total relaxation time [transport time] that should decrease at stronger disorder, i.e. 1/tau ~ n |W|^2 [n is the 2D density of Anderson impurities]. Sure, the ratio of REE is to stay mostly unaffected, but the disorder is still to affect Sy and Jx magnitudes."*

The outcome of the simulations is the calculation of Eqs. (7a) and (7b) of the manuscript, which correspond to the non-equilibrium expectation value (within linear response) of the physical observable represented by some operator *O*, as we do for the current and non-equilibrium spin density. Additionally, our simulations can also compute the spectral expectation value in equilibrium of the observable *O*, which is given by Eq. (12) of the revised manuscript, which was not included in the previous version, as we do for the density of states. **In light of the Reviewer's**

comment, we have modified the Methods section to explicitly state the raw output of our simulations.

The raw output of the simulations for the real-space disordered system is presented in Fig. S2 of the supplementary information. We observe that Sy and Jx are strongly suppressed by disorder outside the pseudogap, whereas the suppression is weaker within the pseudogap. The Reviewer's comment has called our attention to the misleading presentation of these results in the original manuscript (lines 245-254 therein, where it is said "[...] both Sy and Jx remain mainly unaffected by disorder within the pseudogap, while they are suppressed outside [...]"). **We have therefore rewritten the presentation of these results in the revised version**. We note that the simulation's results have not changed between the original and revised manuscript, only the wording has.

> *"5. Strictly speaking, Eq.5 is not actually a clean limit: 5a is nothing but the Drude current which diverges at vanishing scattering (i.e. in the true clean limit). My understanding is that Eq.4 and 5 based analysis corresponds to a simplified treatment of disorder: The scattering time is simply defined by the broadening of the single-particle Green's function poles rather than by self-consistent evaluation from the vertex equations. This, however, does not transform the diffusive transport regime from Eq.5b, so saying "beyond the clean limit" in line 231 [and other places] might be misleading."*

We thank the Reviewer for their comment, as it allows us to better explain the methodology and approximations used in order to obtain Eq. (5) of the manuscript. As the Reviewer correctly understands, in this framework the effects of scattering are only represented by defining a finite energy broadening in the single particle Green's function, while the Hamiltonian and current operators inserted in the Kubo formula are considered to be disorder-free. **We have therefore rephrased this explanation in the revised manuscript, removing the mention to a "clean limit" and replacing it by the "constant broadening approximation" (as is frequently referred to in the literature, or "constant Gamma approximation")**.

> *"6. Is it true that the definition of the REE efficiency [line 188] has an "intrinsic" character: It reflects the intrinsic degree of the spin conversion from the Drude current to the nonequilibrium spin polarization? Does this "intrinsic" quantity actually determine the "external" effectiveness of REE and some related effects [e.g. spin-orbit torques in a neighboring magnet]? Would the usage of the drift*

velocity in line 188 [instead of the Fermi velocity] better reflect the actual amount of spin polarization we can generate and use in real devices?"

The Reviewer proposes an interesting question regarding the most accurate way to characterise the efficiency of the REE. We now argue in favour of the definition employed in the manuscript from a different perspective. In the REE a non-equilibrium spin density is generated due to the out-of-equilibrium electron distribution under an applied electric field, along with an odd-in-momentum spin texture. This out-of-equilibrium electron distribution additionally generates a longitudinal electric current. Therefore, we may quantise the REE efficiency by taking the ratio between the non-equilibrium spin density S_y (normalised by hbar/2) and the non-equilibrium carrier density which contributes to the electric current n_{neq}. The provisional REE efficiency definition is $\Theta_{REE}=(2S_y/\hbar)/n_{neq}$. We now obtain n_{neq} from the longitudinal current. Under the Drude transport model, the drift velocity formulation of the current considers a carrier density defined in equilibrium n_{eq}, which is accelerated by the applied electric field, yielding the current density $j=e*n_{eq}*v_{drift}$, with v_{drift} the drift velocity. Notice that in this model the drift velocity is proportional to the applied electric field. Therefore, using the drift velocity for the REE efficiency, as the Reviewer suggests, would yield an electric-field-dependent REE efficiency which cannot characterise the efficiency of spin generation per unit of electric current, and it is hence discarded by the authors.

Alternatively, one may consider a set of electron Bloch states with well defined velocities in equilibrium, derived from the band structure, where the applied electric field favours the population of electrons with an anti-parallel velocity. This latter perspective corresponds to the semi-classical Boltzmann transport regime under the constant relaxation time approximation, which is equivalent to the Kubo-Bastin approach with a constant energy broadening approximation, as developed in the manuscript. Therefore, it is from this latter approach that one must obtain the non-equilibrium carrier density involved in the REE efficiency, by appropriately normalising the longitudinal current by the Fermi velocity. The mathematics for the Boltzmann framework is more complicated than for the Drude one and is omitted in this answer, but it is indeed present in the manuscript (see Eq. (5b) and the Methods section).

The previously exposed reasoning is already present in Ref. [Offidani, et al. *Phys. Rev. Lett.* **119** 196801 (2017)], where the REE efficiency is first proposed. However, said definition, based on the Fermi velocity, can only be applied to systems where the

band structure is well defined, and therefore does not treat the disorder potential on equal footing to the periodic Hamiltonian. In our work, we extend this definition in order to obtain a *transport velocity* which can be computed in arbitrarily disordered systems, and converges to the band structure Fermi velocity in the limit of a vanishing disorder potential. This allows us to study the REE efficiency in a real-space disordered system with a non-perturbative treatment of disorder.

REVIEWER #3: point-by-point response

"This study reports the emergence of a gigantic current-induced spin polarization in graphene due to spin-pseudospin entanglement control and the interplay between Bychkov-Rashba and Kane-Mele spin-orbit coupling (SOC).

This manuscript is based on an interesting observation: a Kane-Mele spin-orbit coupling (SOC) does not eliminate an important property of the Dirac-Rashba Hamiltonian, i.e., the property that spin-pseudospin four-band Hamiltonian effectively decouples in two two-band Hamiltonians, where the spin and pseudospin wind with the same or opposite sign, respectively. Such a decoupling allows to show that the spin and the pseudospin, are maximally entangled at low energies. It is exactly in this regime that the Kane-Mele SOC partially removes such an entanglement and allows the possibility of maximum efficiency in the spin-charge interconversion. Having established this possibility, the manuscript then systematically explores the Rashba-Edelstein effect and also the spin Hall effect as a function of the Fermi energy, both in the pure and disordered cases. For the latter, they use the Kernel Polynomial Method to study the effect of disorder, introduced via the Anderson model with an onsite random term. The results are interesting and the paper is well written. However, before recommending publication, we would like to draw the authors' attention on the following points, which, if properly addressed, may improve the clarity of the paper."

"1. In Eqs.(5.a-b) the authors report the expressions for the spin density and the charge current density with allowance of a relaxation time τ_η. The authors are right in saying that both physical observables are Fermi surface phenomena, but they should be more clear about the role of the relaxation time. In clean systems both observables are infinite (they scale as the relaxation time), so the introduction of the relaxation time τ_η can be seen as a useful mathematical device to get the finite ratio of two infinite quantities. Therefore, τ_η should not be intended as an effort to study the effect of random potentials in a disordered system, as it would

lead to well known problems related to the absence of vertex corrections in the Kubo formula. The text should make clear this point. We would also point out the validity of your method by saying that the response you obtain with the proper inclusion of disorder produces a response which behaves similarly to the clean one (suggesting that the introduction of a constant relaxation time did not introduce artifacts).”

The Reviewer’s comment is indeed correct, as in a clean system both Fermi surface transport quantities are infinite. The relaxation time τ_η emerges from the introduction of a constant spectral broadening to the Green’s function in an otherwise clean system, which is formally equivalent to the semiclassical Boltzmann transport framework with the constant relaxation time approximation [Zelezny, et al. *Phys. Rev. Lett.* **119**, 187204 (2017)]. Such treatment, as the Reviewer well comments, does not rigorously capture the effects of scattering on random disorder potentials, as it ignores the vertex corrections in the Kubo formula. **In the revised manuscript we have included this analysis on the interpretation and limitations of this methodology. Additionally, to avoid confusion with a true clean limit where the conductivity diverges, we refer to this transport framework as the *constant broadening approximation*.**

The Reviewer also suggests us to analyse to a deeper extent the comparison between the simplified constant broadening approximation with the non-perturbative treatment of disorder in the real-space simulations. We thank the Reviewer for their suggestion, as it improves the delivery and impact of our results. **We have modified the manuscript accordingly, better explaining the relation between both transport frameworks, both respect to the methodology and the obtained results**.

“2. Another issue, partially related with the previous point: Eqs.(5.a-b) are strictly valid only within the pseudogap up to edges or are valid also when two bands cross the Fermi energy?”

Eqs (5.a,b) are valid in both regimes, as they consider a sum carried over all Fermi momenta. Within the pseudogap the sum comprises a single term, as only one band intersects the Fermi level; whereas outside the pseudogap the sum is carried over all band intersections with the Fermi level.

“3. In page 3, the authors discuss the conserved quantity Q. Here, appear the matrices σ_k, σ_ϕ, sk and s_ϕ, which have not been clearly defined before. They also

appear in the Supplementary Note 1 in Eqs.(S3.a-b) without an explicit definition. Of course it is not difficult to deduce it from the context, but certainly it will help the reader to have their explicit definition, and also by stating, for instance, that the asymptotic states at large momenta are the eigenstates of σk. It may be useful to show explicitly the form of the asymptotic eigenstates in the Supplemental Material."

Following the Reviewer's suggestion, we have included an explicit definition for the matrices as well as their relation to the asymptotic states.

"4. In the caption of Fig. 1, while describing panel (a), a yellow arrow is mentioned, but there is not a yellow arrow."

In Fig. 1 (a), of the original manuscript there is a yellow arrow inserted at the center of the helical Fermi contours depicted in the bottom. Nevertheless, the Reviewer's concern made us realise that the depiction of two separate sets of Fermi contours (top, solid and bottom, faded, with yellow arrow) within the same panel is confusing. **We have therefore modified the figure to separate the former panel (a) into two panels (labelled (a) and (b) in the revised manuscript), and modified the caption accordingly.**

"5. In Fig. 2 the red and gray curves, both the full and dashed ones, are related to the different values of the Kane-Mele SOC? But which is which?"

In Fig. 2, all panels, the color of the curves is related to the values of the Kane-Mele SOC according to the colorbar therein. The Reviewer's comment has made us realise that the colorbar jurisdiction over panels (b) and (c) was not clear in the original manuscript. **We have therefore made it explicit in the caption in the revised manuscript.**

"6. In Eq. 2, It may be useful add a symbol indicating the identity matrix in front of the first term on the right hand side of the equation."

We have modified Eq. (2) accordingly in the revised manuscript.

"7. In Eq. 4 the authors introduce the Hamiltonian as "H" for the first time, because, presumably, they apply the Kubo formula either to the reciprocal space low energy Hamiltonian (cf. Eq.(2)) or to the real-space Hamiltonian (cf. Eq. (10)),

so H can be either of the two. It would be useful to say this in the main text, as the reader refers only to the Hamiltonian in Eq. (1) denoted as Hk. As for the clean system calculation, for a single valley, it may be useful to point out that the Hamiltonian to be considered in Eq. (4) is the full 4x4 Hamiltonian with both χ sectors."

We thank the Reviewer for their comment, as it motivates us to explain more clearly our methodology for numerical computations. We perform transport calculations under two regimes. In the constant broadening approximation (as labelled in the revised manuscript, previously referred to as "clean limit") we use the Bloch representation of the Hamiltonian (Eq. (1)) and compute the Kubo-Bastin formula according to Eqs (8) and (9). On the other hand, in the KPM-based simulations we use the real-space tight-binding formulation of the Hamiltonian (Eq. (10)) and compute the Kubo-Bastin formula according to Eqs.(7a) and (7b). **In the revised manuscript we have modified the Methods section, clearly stating the output of our simulations, and in which representation of the Hamiltonian is used. Additionally, we have included the specific mention of "Bloch Hamiltonian" in Eq. (1) to avoid confusions between both representations**.

REVIEWER #4: point-by-point response

"I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Communications Physics 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 participating in the peer review process.

Summary of modifications to the manuscript

- In the Introduction section, in the second paragraph of page 2, we have included the electric dipole spin resonance phenomenon as an example of spin-pseudospin driven physics in graphene, following the suggestion by Reviewer 2.
- When introducing Eq. (1), in page 2, we have explicitly stated that it is the Bloch representation of the Hamiltonian, in order to differentiate the Hamiltonian considered for the kubo-Bastin formula, following the concern of Reviewer 3.

- In page 2, we have modified the discussion on the material platforms which may potentially realise the proposed system. The revised discussion is more clear as to the values for SOC parameters reported in different proximitised graphene systems, following the discussion with Reviewer 2.
- In page 2, we have introduced an explicit definition for the σ_k and s_ϕ matrices, following the suggestion by Reviewer 3.
- In Eq. (2), in page 3, we have explicitly included the identity matrix, following the suggestion by Reviewer 3.
- In page 4, before obtaining the results contained in Eqs. (5a) and (5b), we have modified the description of the transport framework used. While in the previous manuscript this was referred to as the *clean limit*, the Reviewers' remarks pointed out that this name is misleading. We have therefore reformulated the description of this transport framework, to which we refer to as the *constant broadening approximation*, as is also found in the literature. This modification is observed throughout the entire manuscript as well.
- In page 4, we have modified the introduction to the framework for real-space transport calculations in disordered systems. The modified manuscript highlights the difference in the treatment of disorder between both frameworks, and allows us to better analyse our results.
- In page 4, we have modified the analysis of the Rashba-Edelstein effect results in the disordered system, following the comments by Reviewer 2. The modified analysis directly references the raw calculations, and describes more precisely their response to disorder.
- When analysing the results for the spin Hall effect, in page 5, we have emphasised the difference between both treatments of disorder, following the suggestion by Reviewer 3.
- In the Methods section, we have more clearly indicated the raw output of our calculations, as well as the inputs and formulas required for them, following the Reviewers' questions.
- In Fig. 1, we have separated the former panel (a) into two panels, (a) and (b), following the concerns by Reviewer 3.
- In Fig. 2, we have clearly indicated the link between the colourbar and the Kane-Mele SOC value, following the question by Reviewer 3.
- In the supplementary information, we have included the transport results considering a finite temperature, following the question by Reviewer 2.

Report on *Optimal spin-charge interconversion in graphene through spin-pseudospin entanglement control* by Dueñas et al.
COMMSPHYS 25 1672

This study reports the emergence of a gigantic current-induced spin polarization in graphene due to spin-pseudospin entanglement control and the interplay between Bychkov-Rashba and Kane-Mele spin-orbit coupling (SOC).

This manuscript is based on an interesting observation: a Kane-Mele spin-orbit coupling (SOC) does not eliminate an important property of the Dirac-Rashba Hamiltonian, i.e., the property that spin-pseudospin four-band Hamiltonian effectively decouples in two two-band Hamiltonians, where the spin and pseudospin wind with the same or opposite sign, respectively. Such a decoupling allows to show that the spin and the pseudospin, are maximally entangled at low energies. It is exactly in this regime that the Kane-Mele SOC partially removes such an entanglement and allows the possibility of maximum efficiency in the spin-charge interconversion. Having established this possibility, the manuscript then systematically explores the Rashba-Edelstein effect and also the spin Hall effect as a function of the Fermi energy, both in the pure and disordered cases. For the latter, they use the Kernel Polynomial Method to study the effect of disorder, introduced via the Anderson model with an onsite random term. The results are interesting and the paper is well written. However, before recommending publication, we would like to draw the authors'attention on the following points, which, if properly addressed, may improve the clarity of the paper.

1. In Eqs.(5.a-b) the authors report the expressions for the spin density and the charge current density with allowance of a relaxation time τ_η. The authors are right in saying that both physical observables are Fermi surface phenomena, but they should be more clear about the role of the relaxation time. In clean systems both observables are *infinite* (they scale as the relaxation time), so the introduction of the relaxation time τ_η can be seen as a useful mathematical device to get the *finite* ratio of two infinite quantities. Therefore, τ_η should not be intended as an effort to study the effect of random potentials in a disordered system, as it would lead to well known problems related to the absence of vertex corrections in the Kubo formula. The text should make clear this point. We would also point out the validity of your method by saying that the response you obtain with the proper inclusion of disorder produces a response which behaves similarly to the clean one (suggesting that the introduction of a constant relaxation time did not introduce artifacts).

2. Another issue, partially related with the previous point: Eqs.(5.a-b) are strictly valid only within the pseudogap up to edges or are valid also when two bands cross the Fermi energy?

3. In page 3, the authors discuss the conserved quantity $\mathcal{Q}$. Here, appear the matrices σ_k, σ_ϕ, s_k and s_ϕ, which have not been clearly defined before. They also appear in the Supplementary Note 1 in Eqs.(S3.a-b) without

an explicit definition. Of course it is not difficult to deduce it from the context, but certainly it will help the reader to have their explicit definition, and also by stating, for instance, that the asymptotic states at large momenta are the eigenstates of σ_k. It may be useful to show explicitly the form of the asymptotic eigenstates in the Supplemental Material.

4. In the caption of Fig. 1, while describing panel (a), a yellow arrow is mentioned, but there is not a yellow arrow.

5. In Fig. 2 the red and gray curves, both the full and dashed ones, are related to the different values of the Kane-Mele SOC? But which is which?

6. In Eq. 2, It may be useful add a symbol indicating the identity matrix in front of the first term on the right hand side of the equation.

7. In Eq. 4 the authors introduce the Hamiltonian as "H" for the first time, because, presumably, they apply the Kubo formula either to the reciprocal-space low energy Hamiltonian (cf. Eq.(2)) or to the real-space Hamiltonian (cf. Eq. (10)), so H can be either of the two. It would be useful to say this in the main text, as the reader refers only to the Hamiltonian in Eq. (1) denoted as $\mathcal{H}_{\mathbf{k}}$. As for the clean system calculation, for a single valley, it may be useful to point out that the Hamiltonian to be considered in Eq. (4) is the full 4x4 Hamiltonian with both χ sectors.

Second report on *Optimal spin-charge interconversion in graphene through spin-pseudospin entanglement control* by Dueñas et al.
COMMSPHYS 25 1672

We think that the authors have done a good job in the revision of the first version of the manuscript, especially clarifying points which could have been interpreted in an ambiguous way. Furthermore, all the previous criticisms have been convincingly discussed. We then recommend publication. Finally, but this is just an estetic remark, the *yellow arrow* in panel Fig. 1(b) is not obviously evident (A yellow thin line is the contour of a bulk blue big arrow). We leave this to the typesetting office to judge.