Reply to “Comment on ‘Spin-dependent electron transmission model for chiral molecules in mesoscopic devices’”

Xu Yang,* Caspar H. van der Wal, and Bart J. van Wees

Zernike Institute for Advanced Materials, University of Groningen, NL-9747AG Groningen, The Netherlands

(Received 6 November 2019; published 17 January 2020)

In this Reply, we emphasize once more the distinction between generating CISS (spin-charge current conversion) in a chiral spin-orbit system and detecting it as magnetoresistance in two-terminal electronic devices. We also highlight important differences between electrical measurement results obtained in the linear response regime and those obtained in the nonlinear regime.

DOI: 10.1103/PhysRevB.101.026404

The Comment by R. Naaman and D. H. Waldeck [1] addresses our recent publication “Spin-dependent electron transmission model for chiral molecules in mesoscopic devices” [2]. We believe that the Comment is largely based on misunderstandings of Ref. [2], and it is important to clarify these in detail. Therefore, we provide here a point-by-point reply to the Comment and emphasize the important distinctions between the results obtained in the linear response regime and those obtained in the nonlinear regime, because both are experimentally observed using two-terminal electrical measurements. Moreover, we emphasize again the importance of differentiating the conditions for generating a spin polarization in a chiral molecule from those for detecting it as a magnetoresistance signal in a two-terminal electronic device.

(i) Comment: “The paper published by Yang et al. [2] models spin transmission through chiral molecules in mesoscopic devices. Based on their model, they claim that spin selectivity in electron transport through chiral molecules, in the linear regime, cannot be measured by using a two-terminal device, unless a spin-flip process occurs in the molecule.”

Our remark: How a spin polarization can be generated by a chiral molecule has indeed been discussed in many publications, including Refs. [3–5] mentioned in the Comment. This is also pointed out in Ref. [2]. However, Ref. [2] addresses a completely different issue, which is how such a spin polarization can be detected as a charge signal in transport experiments in the linear response regime. To our best knowledge, this issue is only addressed in one other publication [6], which appeared after Ref. [2].

(ii) Comment: “Their simplified, two-terminal model assumes that charge is injected from a source electrode, transits through a chiral molecule and a ferromagnet, and is collected at a drain electrode. In this treatment, the ferromagnet transmits a given spin and reflects the other; but there is no dissipation in the ferromagnet. While the conclusions drawn by the authors may be consistent with the simplified model, the model itself is not realistic enough to account for experiments.”

Our remark: We indeed consider a two-terminal model but it does not involve simplifications for the linear response regime. Note that the role of the source and drain electrodes are interchangeable in the linear response regime because of microscopic reversibility. We have included in the model electron reflections at all interfaces, including the ferromagnet and the electrodes. The ferromagnet is characterized by a spin polarization parameter, which can be tuned from 0–1. The conclusions of the model are valid for all polarization values.

(iii) Comment: “Theoretical models for the chiral-induced spin-selectivity (CISS) effect, in two-contact spin measurements, exist in the literature already, and the conditions for observing spin polarization have been discussed in detail. As an example, consider the work by Matityahu et al. [3] which states: ‘When the helix is connected to two one-dimensional single-mode leads, time-reversal symmetry prevents spin polarization of the outgoing electrons. One possible way to retrieve such a polarization is to allow leakage of electrons from the helix to the environment, via additional outgoing leads.’”

Our remark: How a spin polarization can be generated by a chiral molecule has indeed been discussed in many publications, including Refs. [3–5] mentioned in the Comment. This is also pointed out in Ref. [2]. However, Ref. [2] addresses a completely different issue, which is how such a spin polarization can be detected as a charge signal in transport experiments in the linear response regime. To our best knowledge, this issue is only addressed in one other publication [6], which appeared after Ref. [2].

(iv) Comment: “In other words, dephasing acts to create asymmetry in the transmission amplitude for spin up versus spin down, and it breaks Onsager’s reciprocity relation.”

Our remark: While dephasing indeed creates transmission asymmetry for opposite spins, it does not break reciprocity. The Onsager’s relation is a thermodynamical theorem, and it holds in the presence of dephasing, see, for example, Ref. [7].

(v) Comment: “For example, Sánchez and Buttiker [8] showed how asymmetry arises for magnetoresistance in a two-terminal device. The combination of interactions with a bath and the large electric fields at interfaces (typical of CISS experiments) can result in the observed asymmetry.”

Our remark: This is correct, but the asymmetry can only occur outside the linear response regime, i.e., away from zero bias by at least $V = k_B T / e$ [9].
COMMENTS

Our remark: This is the same issue as explained in the above Point (iii). References [4,5] discuss how a spin-backscattering, as an explanation for the spin selectivity, was also discussed previously [4] and even used to analyze for the extent of spin flipping in experiments [5].

Our remark: This is irrelevant to the linear response regime. The spin-backscattering, as an explanation for the spin selectivity, was also discussed previously [4] and even used to analyze for the extent of spin flipping in experiments [5].

Our remark: This description is not consistent with our model. According to Ref. [2], the polarization calculated as in Fig. 1(c) should be zero in the linear response regime, and then it may increase with increasing bias. Both curves in Fig. 1(c) indeed show zero spin polarization at zero bias, which proves the relevance of our model to actual measurements.

(xi) Comment: “In summary, the model presented in Ref. [2] oversimplifies; it fails to include the dissipation processes occurring at room temperature and it considers a linear limit that is not valid for the measurements on the CISS effect.”

Our remark: This conclusion is incorrect. Reference [2] considers only the linear response regime, which is clearly observed in experiments such as the one shown in Fig. 1.
in the Comment, and therefore it is very relevant to actual measurements. Within the linear response regime the model is not simplified.

We emphasize again that Ref. [2] is intended to raise awareness of the consequences of fundamental symmetries and limitations of certain electrical measurement geometries. It also highlights the differences between linear and nonlinear regimes. An extension of our model shows that a magnetoresistance in the two-terminal geometries discussed here can indeed be observed in the nonlinear regime [14].

[1] R. Naaman and D. H. Waldeck, Comment on “Spin-dependent electron transmission model for chiral molecules in mesoscopic devices”, Phys. Rev. B 101, 026403 (2020).
[2] X. Yang, C. H. van der Wal, and B. J. van Wees, Spin-dependent electron transmission model for chiral molecules in mesoscopic devices, Phys. Rev. B 99, 024418 (2019).
[3] S. Matityahu, Y. Utsumi, A. Aharony, O. Entin-Wohlman, and C. A Balseiro, Spin-dependent transport through a chiral molecule in the presence of spin-orbit interaction and nonunitary effects, Phys. Rev. B 93, 075407 (2016).
[4] E. Medina, L. A. González-Arraga, D. Finkelstein-Shapiro, B. Berche, and V. Mujica, Continuum model for chiral induced spin selectivity in helical molecules, J. Chem. Phys. 142, 194308 (2015).
[5] D. Nürenberg and H. Zacharias, Evaluation of spin-flip scattering in chirality-induced spin selectivity using the riccati equation, Phys. Chem. Chem. Phys. 21, 3761 (2019).
[6] S. Dalum and P. Hedegård, Theory of chiral induced spin selectivity, Nano Lett. 19, 5253 (2019).
[7] M. Büttiker, Role of quantum coherence in series resistors, Phys. Rev. B 33, 3020 (1986).
[8] D. Sánchez and M. Büttiker, Magnetic-Field Asymmetry of Nonlinear Mesoscopic Transport, Phys. Rev. Lett. 93, 106802 (2004).
[9] Supriyo Datta, Electronic Transport in Mesoscopic Systems (Cambridge University Press, Cambridge, 1997).
[10] M. Ben-Chorin, F. Möller, and F. Koch, Nonlinear electrical transport in porous silicon, Phys. Rev. B 49, 2981 (1994).
[11] A. A. Middleton and N. S. Wingreen, Collective Transport in Arrays of Small Metallic Dots, Phys. Rev. Lett. 71, 3198 (1993).
[12] K. Michaeli, D. N. Beratan, D. H. Waldeck, and R. Naaman, Voltage-induced long-range coherent electron transfer through organic molecules, Proc. Natl. Acad. Sci. USA 116, 5931 (2019).
[13] V. Kiran, S. P. Mathew, S. R. Cohen, I. Hernández Delgado, J. Lacour, and R. Naaman, Helicenes—a new class of organic spin filter, Adv. Mater. 28, 1957 (2016).
[14] X. Yang, C. H. van der Wal, and B. J. van Wees, Detecting chirality in two-terminal electronic devices, arXiv:1912.09085.