June 16, 2023

Re: XW10765L
Transport of Spin Magnetic Multipole Moments Carried by Bloch Quasiparticles

We thank both referees for carefully reading through our manuscript and response files. We are delighted to see that all referees have expressed their recognition of the significance and novelty of our work. We hope our detailed response below together with the revised manuscript can fully address the remaining concerns of the referees, and our paper can now be accepted for publication in PRL.

Summary of major changes in the revision:

1. We have rewritten the introduction section to incorporate the key points in our previous response regarding the importance of our work.

2. We have added a new subsection in Sec. I of the supplementary material to give an alternative and physically more intuitive derivation of the multipole moment formula.

3. We have added a new paragraph in the discussion section on the dependence of wave-packet shape in higher-order semiclassical theories.

Second Report of Referee B -- XW10765L/Tahir

Comment: *The authors have done serious work on the manuscript. I am also impressed with their detailed response to my comments. After studying this response, I am ready to believe that the topic under study is really of interest to a wide range of readers. In principle, the paper could be published in PRL. However, I am not entirely satisfied with the authors' answers to some specific questions I asked:*

Reply: We are encouraged to hear that the referee now appreciates the significance and novelty of our work. We hope our detailed responses below can address their remaining concerns and our paper can be recommended by the referee for publication in PRL.

Question #1: *The authors propose a specific version of the formula that describes the multipole moment. This formula is gauge invariant and does not depend on the position of the center of mass (formula 4 of the corrected version). I asked in the first round of peer review about the status of this formula. I have carefully read their response to my remark, but still do not understand whether this formula is strictly derived or whether the authors offer a plausible answer. In more detail, the starting formula (SM 11) in Supplemental Material (SM) is not yet gauge invariant, as the authors demonstrate in the example (SM13). At the same time, formula (SM 20) is already gauge invariant. After reading the text between (SM13) and (SM20), I got the impression that the authors simply replaced the $i d_{kj}$ derivatives with the "long" derivatives $ i d__{k_i} - A_i.$ If so, it should be written explicitly. Although such a procedure is reasonable, it is not fully justified. Some physical justification is required. In addition, the question of the uniqueness of such a procedure arises. As far as I understand, $i d__{k_x} - A_x$ and $i d__{k_y} - A_y$ generally do not commute, so the question arises about the correct order of these operators. It still seems to me that the authors should provide more detailed explanations on this issue.*

Reply: We thank the referee for critically reading through the previous version of our manuscript files. Before going into the details, we point out that, rigorously speaking, our wave-packet multipole moment formula relies on choosing the center-of-mass position of the wave packet as the origin, and is therefore more of a definition rather than derived from first principles. Nonetheless, it is expected that when a natural separation of length scales applies, such as that between the wave-packet size (e.g., the Compton wavelength of Dirac electrons) and the mean-free path, such a gauge-invariant quantity can be exposed by specifically designed experiments probing the relevant length scales.

To provide a more illuminating justification for our formula Eq. (4), we have added an alternative derivation of it as a new subsection in Sec. I of the revised SM and mentioned it below Eq. (5) in the main text. Here we summarize the central idea which is based on canonical quantization of semiclassical wave-packet theory (Refs. [81,86]).

The gauge-dependent issue with the naïve definition of the wave-packet multipole moment

$$\mathcal{M}^{i_l}_{i_1 i_2 \ldots i_{l-1}} \equiv \langle W| \prod_{n=1}^{l-1} (\mathbf{r}-\mathbf{r}_c)_{i_n} s_{i_l} |W\rangle$$

is fundamentally caused by the fact that $\mathbf{r}_c$ being a c-number does not have a well-defined value. In contrast, the center-of-mass crystal momentum $\mathbf{k}_c$ of the wave packet is always well defined. Therefore, as a minimal extension of the above formula, one can try to introduce $\mathbf{r}_c$ through $\mathbf{k}_c$. This can be done by promoting the pair of semiclassical variables $\mathbf{r}_c$ and $\mathbf{k}_c$ to re-quantized operators $\hat{\mathbf{r}}_c$ and $\hat{\mathbf{k}}_c$ acting on a new

Hilbert space spanned by the wave packet states $\{|W_{\mathbf{k}}\rangle\}$, which are eigenstates of $\hat{\mathbf{k}}_c$. One can then show that

$$\hat{\mathbf{r}}_c|W_{\mathbf{k}}\rangle = (-i\partial_{\mathbf{k}} + \mathcal{A}_{\mathbf{k}})|W_{\mathbf{k}}\rangle$$

where the $\mathbf{k}$ derivative is due to the canonical conjugate of $\hat{\mathbf{k}}_c$, $\hat{\mathbf{r}}_c^{\mathrm{can}}$, that satisfies $[\hat{\mathbf{r}}_c^{\mathrm{can}}, \hat{\mathbf{k}}_c] = i$. The appearance of the Berry connection is very much analogous to that of the vector potential in Peierls substitution, which is the difference between physical and canonical momenta. Similarly, due to the presence of the Berry connection, different components of $\hat{\mathbf{r}}_c$ do not commute and symmetrization must be done when their products appear in canonical quantization to ensure hermiticity. Then we can start with the following definition of $\mathcal{M}$ modified from the above naïve formula

$$(\mathcal{M}_{n\mathbf{k}_c})^{i_l}_{i_1 i_2 \dots i_{l-1}} \equiv \frac{1}{(l-1)!}\mathrm{Re}\int_{\mathrm{BZ}} d^3\mathbf{k}\langle W_{\mathbf{k}}|s_{i_l}\prod_{m=1}^{l-1}(\mathbf{r}-\hat{\mathbf{r}}_c)_{i_m}|W_{\mathbf{k}_c}\rangle + (\{i_m\}\text{ permutations})$$

where the critical difference is to replace $\mathbf{r}_c$ by $\hat{\mathbf{r}}_c$, to arrive at Eq. (4) in the main text after some algebra.

The above heuristic approach is a nontrivial result by itself since it does not require a separate treatment to eliminate the wave-packet shape dependence and also points out a route for generalizing the multipole moment formula to the case of degenerate bands, which will be reported in a future work. The original approach is still necessary since it gives a directly computable formula through elementary calculations and proves its gauge invariance.

Question #2: *I am also not completely satisfied with the authors' answer to the question of what happens to formula (11) (new version) at $\Lambda^R_0 \to 0$. As far as I understand the text including Eqs. (SM93) and (SM94), the authors state the following: (i) the contributions from K_v and $ - K_v$ cancel each other out exactly (ii) even when limited to only the contribution from $K_v,$ it vanishes if $ \Lambda^R_0 \to 0$ when taking into account the higher conductive band n=3. This means that there is some characteristic value of $\Lambda^R_0, $ starting from which the contribution from the zone n=3 comes into play. From the written text, this parameter is not clear. The authors must provide some kind of estimate of this parameter.*

Reply: The reason why we brought up the higher conduction band is that macroscopic observables, such as the current-induced multipole moment [Eq. (7) in the main text], only depends on the sum of the contributions from different bands, weighted by their respective distribution functions. As $\Lambda_0^R \to 0$, the two conduction bands become nearly degenerate, so that they have nearly the same coefficients of the distribution function. Moreover, the area in the Brillouin zone in which the net contribution is nonzero also shrinks. As a result, the macroscopic octupole moment has no singular behavior as $\Lambda_0^R \to 0$.

To demonstrate the above statements, we show in the figure below the integrand of Eq. (7) in the main text plotted along the $k_y = 0$ line near the Fermi surface for different sizes of Λ_0^R relative to the original value used in the main text. One can see that for each curve, there are two peaks with opposite signs coming from the Fermi surfaces of the two conduction bands, respectively. The peak values decrease as Λ_0^R increases due to the inverse proportionality between $\mathcal{M}$ and Λ_0^R as the referee noticed. However, the integrals of the three curves in the field of view have much smaller values: -9.58, -9.63, and -9.82 for Λ_0^R equal to 1, 5, 10 times the original value, respectively, in the units of the integrand divided by Å. This is partly due to the narrowness of the peaks in momentum space, and partly due to the opposite signs of the contributions from the two bands, as explained above. Moreover, the integrated value increases with increasing Λ_0^R, which is reasonable since larger Rashba spin-orbit splitting should generally enhance the observed current-induced spin magnetic multipole moments.

We would like to further elaborate upon the following point in our previous answer to the referee's same question: The behavior of individually diverging but mutually cancelling contributions from near-degenerate bands is similar to that of Berry curvature as the integrand of the intrinsic anomalous Hall conductivity. The Berry curvature of a given band diverges as the energy difference between it and another band at the same **k** approaches zero. However, the Berry curvature of the latter band also diverges for the same reason but has an opposite sign. As a result, after summing over occupied bands, nonzero contributions of the Berry curvature to the integration only come from narrow regions in the Brillouin zone where the near-degenerate bands cross the Fermi energy [see, e.g., PRL 92, 037204 (2004) and PRB 74, 195118 (2006)]. Although in these regions the Berry curvature summed over occupied bands can be gigantic, its integration is still regular.

In the revised supplementary material we have added the following sentence to the end of the paragraph below Eq. (109) on page 16:

As a result, macroscopic values of the multipole moments after Brillouin zone integration of the individual bands' contributions weighted by the distribution function do not have singular behavior, similar to the case of intrinsic anomalous Hall conductivity.

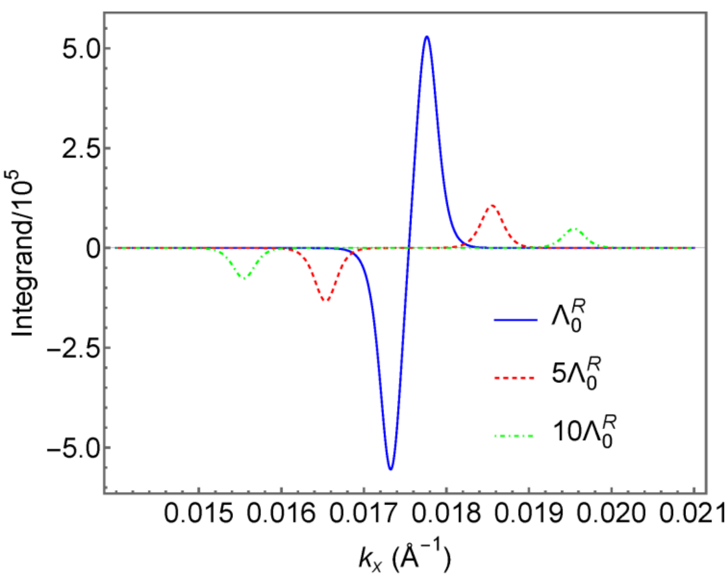

Fig. R1. Integrand of Eq. (7) of the main text for $\mathcal{M}_{xy}^{x}$ (divided by 10^5 for easier visualization) with $E_F - \Delta = 0.01$ eV, $k_B T$ =0.1 meV, and **E** $\parallel \hat{x}$, plotted along $k_y = 0$ for different values of Λ_0^R relative to the original value used in the main text.

Question #3: *The authors have provided a sufficiently detailed justification of the importance of the topics studied in response to the criticism of both referees. I would like something from this answer to be presented in the main text of the paper.*

Reply: We thank the referee for this suggestion. We have rewritten the introduction, which now incorporates the central messages in our previous answer.

Report of Referee C -- XW10765L/Tahir

Comment: *On reading the paper and the comments from Referees A and B (as well as the replies) I believe that the authors are writing on a subject that is novel and of sufficiently wide interest for a broad audience in PRL. Coupling to multi-polar moments either electric or magnetic is recently becoming a core issue in a number of areas not least in spintronics that the authors concentrate on but in as disparate fields as nanophotonics and even the fractional quantum Hall effect (the electric quadrupole moment plays a key role in the stabilization of FQHE phases).*

The authors' contribution seems to be focused on a microscopic wave packet analysis of high order magnetic moments (as opposed to thermodynamic arguments) and in that sense can be view as a sort of contribution in the same spirit (and impact as) the pioneering work of Phys. Rev. Lett. 99, 197202 (2007) in magnetic moments. The model demonstration in phosphorene is also neat.

So my recommendation in brief is to publish in PRL. In response to questions posed by the PRL editor: Yes, I think the authors have made sufficient changes to their manuscript and have responded adequately to the referees.

Reply: We appreciate the referee's high evaluation of our work and their recommendations.

Comment*: I would, however, have liked the authors to have commented a little more on the difficulties of wave packet analysis particularly for multipolar moments. While I appreciate that their expression is gauge invariant (and follows from traditional Jackson formula, see reply to Referee B), it has been recently pointed out that there are difficulties with using wave packet analysis to model the transport of particles beyond their simple center of mass motion (see extensive discussion in Phys. Rev. Lett. 126, 156602). For instance, Phys. Rev. Lett. 126, 156602 finds that semiclassics would obtain a *non-universal* wave packet width dependent transport while simple Kubo does not. I do not expect, nor would I recommend that the authors address this fully or even at all in this paper (I think it would distract from their nice discussion already), but at least some acknowledgment of possible issues would help signpost the readers and future workers in the field to this.*

Reply: We thank the referee for bringing this interesting paper to our attention. We agree with the conclusion of this paper, also mentioned by the referee, that the wave-packet approach in general gives results that depend on the shape of the wave packet. On the other hand, macroscopic observables must not depend on the choice of wave packet constructions. Therefore the wave-packet shape dependence in the center-of-mass equations of motion must be compensated by other contributions to the final macroscopic observable, e.g. Hall conductivity, that is measured or calculated for a given system with definite boundary conditions. However, we believe that the gauge-invariant and shape-independent part of quantities derived from a wave-packet approach, which are what we have strived to get in the present work, carry a universal meaning and are most suited to capture transport among spatially separated subsystems. Nonetheless, if through careful experimental designs, Bloch wave packets can be individually prepared and/or detected in ultra-clean systems, the wave-packet shape dependence is physically realistic and should be able to be measured.

In the revised manuscript we have added the following paragraph to the discussion section:

Separately, higher-order semiclassical theories based on the wave-packet approach in general include quantities that depend on the wave packet shape. Such shape dependence must be compensated by other contributions to the final macroscopic observable for a given system, although it can also be potentially exposed by experiments on individually prepared wave packets. In this regard, we expect the shape-independent wave-packet spin multipole moment given in this work to be most suitable for describing diffusive transport among spatially separated subsystems.