Answer to Referees EJP-B

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Dear Editor,

We are submitting a revised version of our manuscript “Negative differential conductivity in liquid aluminum from real-time quantum simulations” for publications in European Physics Journal B.

We thank the reviewers for their comments and suggestion. We have taken all of them into consideration. We include below the answers to each comment, together with the changes we made to the article.

Regards,

Xavier Andrade
Sebastián Hamel
Alfredo Correa

Report from Referee 1

Overall, this is a very nice and very interesting paper which presents beautiful numerical results. The paper will be suitable for publication after the following minor comments have been addressed:

Overall, this is a very nice and very interesting paper which presents beautiful numerical results. The paper will be suitable for publication after the following minor comments have been addressed: \textit{In this paper, the authors present a computational study of the electronic response of liquid aluminum driven by short and intense electric fields. The calculations are done using the octopus code for a periodic system with a large unit cell (256 atoms) and using the Yabana-Bertsch implementation of TDDFT for periodic solids. The system is driven by a “kicked” electric field, and the time-dependent currents are calculated afterwards. It is found that the currents relax exponentially, from which one can then calculate the electric conductivity. Interestingly, the current quickly evolves from quasi-uniform to highly disordered and localized, with a nonvanishing local average current (eventually, of course, coupling with the ions would result in a dissipation of these currents as well). The calculations are then repeated for various field strengths, and it is found that the system does not obey Ohm’s law: instead, a negative differential conductivity is found. The numerical results are compared with theoretical modelling using Lindhard theory. These are novel and very interesting results, which have not yet been experimentally observed, but could be accessible with the next generation of intense light sources.}

Overall, this is a very nice and very interesting paper which presents beautiful numerical results. The paper will be suitable for publication after the following minor comments have
been addressed:
To conform to the Journal style, the paper should be divided into sections.
The paper is now divided in sections.

Figure 4: the caption seems to be cut off.
This was a compilation error. It is fixed in the new version.

In the paragraph after Eq. (2), the authors mention certain pathologies that occur under perfect crystal symmetry. Perhaps add a quick comment as to what these pathologies are, and perhaps include a reference.

The comment is simply related to the fact that electrons would form Bloch waves associated with currents that, due to crystalline translation symmetry, wouldn't decay. We added the sentence “In a perfect crystalline solid, an initial current would produce Bloch waves that support persistent (non-decaying) currents.” and a citation to Kittel’s book.

In Figure 1, the average norm decays exponentially, but around 1 fs it approaches a constant. What determines this time scale?

As we tried to convey, in an energy-conserving simulation (NVE) it is natural to expect that the system reaches some kind of dynamic equilibrium with persistent fluctuations that are in relation to the energy excess with respect to the ground state. (Like a classical molecular dynamics simulation would achieve after the system thermalizes).

The energy added to the system is associated with a certain level of fluctuations associated with non-zero instantaneous local current. Once the system reaches that level it stays. We don’t think the characteristic time or mechanism to reach this point is different from the one associated with the average current decay.

- 3 lines after Eq. (1): ”contains” instead of ”containts
- Caption of Figure 6: ”Dotted” instead of ”Dotter”, and ”simple model, Eq. (3),” instead of ”simple model 3”
- Page 5, left column, line 13: ”tails” instead of ”appendices”
- Page 5, right column, line 2 from below: ”phenomenon”

Some minor points and typos:
- 3 lines after Eq. (1): ”contains” instead of ”containts
- Caption of Figure 6: ”Dotted” instead of ”Dotter”, and ”simple model, Eq. (3),” instead of ”simple model 3”
- Page 5, left column, line 13: ”tails” instead of ”appendices”
- Page 5, right column, line 2 from below: ”phenomenon”

Thank you for the corrections, we have incorporated them.

Report from Referee 2

Nevertheless, some minor comments can be raised:
1. p. 3: The paragraph about time-reversible equations leading a time-irreversible current is not convincing. The final currents are a consequence of the initial kick which breaks the symmetry. Is there something more about it? Please be more specific.

Nevertheless, some minor comments can be raised: The manuscript describes a new application of real-time TDDFT: the evaluation of the conductivity in the high-current regime. There, the usual linear-response is not sufficient and non-linear effects (such as negative differential conductivity) can occur. The paper provides us with a new approach and original results. It should be published in EPJ-B.

Nevertheless, some minor comments can be raised:

1. p. 3: The paragraph about time-reversible equations leading a time-irreversible current is not convincing. The final currents are a consequence of the initial kick which breaks the symmetry. Is there something more about it? Please be more specific.

We agree with the referee, the currents at all times are a consequence of the initial kick, which is causal to the breaking of time-reversal symmetry. This leads to a quantum state that is characterized by complex-valued wavefunctions.

The point of the paragraph is that, despite this persistent time evolution (wavefunction are complex at all times and local current fluctuation are non-zero), the process presents relaxation macroscopically. After the relaxation is established at large times, it is not possible to macroscopically distinguish forward or backward time propagation from the observation of macroscopic quantities.

While this might seem surprising in the context of quantum electrons, in the context of classical molecular dynamics this type of long term evolution is the norm. Microscopic equations and trajectories are time-reversible while large systems still reach equilibrium and there is a well defined arrow of time.

We now removed the sentence that can lead to confusion: “The immediate conclusion is that the final state, although is time-reversible on its macroscopic averages, is still a time-dependent quantum state without time-reversal symmetry.”

2. Fig. 4: Please set the maximum of the y-axis to the number of valence electrons, which is the theoretical maximum for the conducting electron count.

We agree. We have changed the plot in the article.

3. Fig. 6: The units in the y-axis is incorrect.

Yes, the Omega symbol was missing.

4. p. 7: The jellium modeling is very nice. Of course, there is a tuned parameter $R_s$. How sensitive is the curve in Fig. 6 with respect to $R_s$?

The main effect of changes $R_s$ is a vertical scaling of the curve as it controls the range of electrons that each ion interacts with. For example, if $R_s = \infty$ the conductivity curve reduces by a factor 5 in the range displayed, basically reproducing the inverse stopping power of single aluminum atom (normalized) in a Jellium. In summary, the result is sensitive to the value of $R_s$ but it is only adjusted to reproduce the asymptotic divergence of curve.

5. p. 7: Could you clarify more the construction of the Fig. 8?

All the information is extracted from the curve $\sigma(J)$ and its definition. It represent the relation $E = \sigma(J) J$ where the independent variable is $J$. That is, for each current $J$ we deduce what is the electric field necessary to maintain that current. Conceptually is different from the simulation results presented Fig. 6 but operationally doesn’t contain more information.
6. p. 7: typo in "prototypical"

Corrected.