Answer to Referee 3

This work uses real-time simulations of two-dimensional crystals in order to study sum and difference frequency generation as well as second-harmonic generation.

The main contribution of this work is a signal processing strategy that is improved with respect to the previous work Ref. [11] by the same team. Consequently, the core part of the manuscript is its section 3. Given that this is incremental technical progress, I would find the work better placed in SciPost Physics Core.

We thank the referee for their assessment and the time spent in carefully reviewing the manuscript. We respectfully disagree with the view that our work represents only incremental technical progress. We believe that our approach opens a new pathway for investigating nonlinear optical responses in 2D and bulk materials, with clear potential for multi-pronged follow-up studies (a few examples are discussed in the conclusions of our manuscript), that is one of the criteria for SciPost Physics. Moreover, we would also like to point out that there is growing interest in the nonlinear responses obtained from real-time simulations. For example, very recently PRL published an article (PRL 135, 026401) on exactly the same topic as our paper, and honestly their method is not better than ours (we added a reference to this work in the present version of the manuscript).

That being said, there is also the application to monolayer h-BN and MoS2 and second-harmonic generation in bilayer h-BN. In this context, I unfortunately have an issue with the underlying Eq. (2). According to the Hohenberg-Kohn theorem, ground states for a many-body system can be described by a density functional. While I am aware that it is common practice to apply this also to band structures, this is already starting to stretch things. Once one gets to time-dependent density-functional theory (TDDFT) -which is basically the situation investigated here- things get more complicated. Thus, as someone who is not an expert in the physics of excitons, I cannot help wondering if the additional terms in Eq. (2) really go into the right direction from the independent-particle picture. If the authors could add a few further remarks to justify this approximation, this could make the difference to render the manuscript publishable in SciPost Physics.

Regarding "applying DFT to band structures", the Hamiltonian in Eq. (2) (Eq. 4 in the present version of the manuscript) contains quasiparticle corrections which are calculated within the GW approximation, the state-of-the-art to calculate quasiparticles energies from first-principles. Then, our starting point are not the Kohn-Sham eigenvalues, but the quasiparticle energies. Regarding how the excitonic physics is included, at variance with TDDFT, instead of the exchange-correlation potential V_{xc}, the Hamiltonian contains the screened-exchange self-energy Σ_{SEX}. This is a non-local operator which describes the long-range electron-hole interaction screened by the electrons in the system. The screening is calculated from first-principles within the Random-Phase approximation. The Σ_{SEX} corresponds within linear-response to the kernel in the Bethe-Salpeter equation (BSE). Both GW and the BSE are derived within diagrammatic Many-Body perturbation theory that allows to control which physical effects are included (this is not possible with common approximations of the V_{xc} within TDDFT, see e.g. Rev. Mod. Phys. 74, 601). Combined with the DFT for the ground-state, GW+BSE is the state-of-the-art to accurately reproduce optical spectra within linear response. This approach is then well-justified theoretically (through Many-Body techniques) and during the last two decades has been assessed successfully against the experimental optical spectra of a broad range of materials. The Hamiltonian in Eq. (2) corresponds to the GW+BSE, and the corresponding real-time approach gives the same results as GW+BSE in the linear response limit. Since, GW+BSE is state-of-the-art and well-established within the first-principles electronic-structure community and the Hamiltonian has been justified in previous publications, we left out some of the details in the previous version of the manuscript. We agree with the referee that it would be beneficial to better detail the physics behind the Hamiltonian, so to make the manuscript self-contained and more accessible to those not familiar with those approaches. In the present version of the manuscript we included a more detailed and less specialistic discussion of the Hamiltonian.

There are a number of relatively minor details (some of which were also noted by Referee 2) that I list among "Requested changes".

1- Provide some justification of Eq. (2).

In the new version of the manuscript we added a discussion of each single term of the effective Hamiltonian in Eq.2, and discuss its limit in the case of small perturbation.

2- The acronym "DFG" carries two meanings in the present manuscript. I thus recommend that the authors check if they cannot find better acronyms than "SFG" and "DFG".

We removed the "DFG" acronym from the acknowledgments

3- Likewise, "DFT" is usually understood to mean "density-functional theory" and this method is actually used in the present manuscript. Again, I recommend looking for a different abbreviation of "discrete Fourier transform" in order to avoid confusion.

We thank the referee for this comment, now in the text we use "discrete FT" for "discrete Fourier transform"

4- Fig. 2 is referenced and discussed only after Fig. 3. I thus recommend moving it to a later place (where it belongs).

We moved Fig. 2 lower (now Fig. 3) and Fig. 3 (now Fig. 2) higher.

5- The end of the first paragraph of section 3.2.1 refers to Fig. 3. However, I see no logarithmic sampling in Fig. 3. Do the authors maybe mean Fig. 2?

We agree with the referee. We meant Fig. 2 (now Fig. 3) and corrected it.

6- There is a number of minor linguistic and typographic issues. I uploaded an annotated manuscript for the authors to help them with proofreading it.

We thank the referee for these corrections, we fixed all typos reported in the annotated manuscript