In what follows, black text is for extracts from editor communication and referees' reports; blue text for the authors' answers.

Reply to editor communication

We finally have three Reports on your revised version which is actually more than we need. Furthermore, the overall tenor is positive. Nevertheless, one should not forget that it was exceptionally difficult to get Reports. While this should not be a criterion in its own right, it really is exceptional to have to contact 11 Referees in round 1 to get a single Report, and then a further 14 Referees in round 2 to get at least one further Report after more than three months (well, we ended up getting three, but this took almost four months). You might also care to know that two of the four Reports that finally came in are not from the semiconductor community, and neither is the Editor in Charge. Short, one should probably face the fact the semiconductor community is not very much interested. At the same time, the signal analysis proposed in section 3 of your manuscript might be of interest also for problems not described by your Eqs. (1,2). For example, there are well-established approaches to simulate time evolution for a spin-1/2 Heisenberg chain, and then one could use your approach to extract the nonlinear corresponding response functions, couldn't one? It might thus be worthwhile to reconsider title, abstract, and the discussion around Eq. (2) in order to appeal to a broader audience. At the same time, you should of course also address the questions that have been raised concerning h-BN and MoS_2.

We would like to thank the editor for their patience and attention dedicated to our manuscript. We recognize that nonlinear response (and specifically processes such as SFG/DFG) is not very much yet explored within the theoretical/computational condensed matter community. This may have further complicated the already hard task of finding referees. We agree that the method we present has a broader scope and can be applied to other Hamiltonians and response functions. Accordingly, we have changed the title, modified the abstract and slightly amended the manuscript where relevant to highlight the generality of the approach. On the other hand, we prefer to maintain the focus on SFG/DFG, as we consider it to be an important topic in its own right (see also reports of referees 2 and 4). In fact, nonlinear responses, and specifically frequency-mixing techniques as those based on SFG/DFG, can reveal, as shown by our results, interesting exciton physics such as exciton-exciton interaction, which cannot be seen by more standard spectroscopic techniques. Accurate first-principles computational tools are important for guiding and interpreting these experiments. Given that such tools were not available, simulations for these phenomena could not be carried out which contributed to the topic not being very much explored so far. Our manuscript (together with the corresponding open-source code freely available to the community) is addressing this gap. In fact, in the users-forum of the Yambo code, we have received an increasing number of enquiries from researchers attempting to use the code to interpret nonlinear processes such as SFG/DFG. We are thus confident there will be enough attention for the topic of the manuscript in the theoretical/computational condensed matter community.
During the whole reviewing process, the manuscript has been reviewed by four referees. Three referees recommended the publication of the manuscript, recognizing its original contribution and interest to the community e.g.
"The results constitute a valuable contribution to the field of computational non-linear optical spectroscopy. Clearly, this manuscript is suitable for publication in SciPost Physics." (Referee 2),
"I believe the manuscript is an important contribution in the field of nonlinear response of materials. The studied effects are relatively unknown, and the level of the developed theory is clearly cutting edge, as it includes many-body excitonic effects that play an important role in 2D materials like the ones studied in the manuscript. Therefore, I recommend the publication in SciPost Physics." (Referee 4).
Referee 3 considered instead the work as incremental and proposed to transfer our manuscript to SciPost Core. At this regards, note that in the first round of reviews, Referee 1 stated
"It is 'incremental', but that is not negative: it clearly builds on earlier work and takes it one (plausible) step further. This makes it the presentation of a useful tool that may also help others who have to deal with extracting frequency-dependent information from time-dependent data. Also, the applications that the authors discuss are well motivated and illustrative. On sees that it is nice to be able to include electron-hole interactions, at least in some materials. They also suggest that the method can indeed be practically useful for understanding experimental data. The criterion "Detail a groundbreaking theoretical/experimental/computational discovery" can be considered as fulfilled, and one can hope for

"Open a new pathway in an existing or a new research direction, with clear potential for multi-pronged follow-up work".
Here below, we address all the referees queries and in the answer to Referee 3 we argue why we believe our work deserves the attention of SciPost Physics.
We believe that the present version of the manuscript, which includes the suggestions from both the editor and the referees to improve clarity, readability and broaden the scope, is suitable for publication in SciPost Physics.

Answer to Referee 2

[...] The manuscript is very clearly written. The results constitute a valuable contribution to the field of computational non-linear optical spectroscopy. Clearly, this manuscript is suitable for publication in SciPost Physics.

We thank the referee for their assessment and the time spent in carefully reviewing the manuscript.

(1) If the authors would like to improve the discussion of their results on hBN and MoS2, they might consider to add results on the band-structures and on the (linear) optical spectra. Even though these are pretty well known in the literature, they could be used to visualize the discussion of the computational results. This might improve the readability and clarity of the second-order results in this manuscript.

We follow the suggestion of the referee and added a new figure (Fig. 5) with the linear optical spectra for hBN and MoS2.

(2) Apart from this, I have only a few typos that should be corrected: - page 3, second line from bottom: "can be accounted for" (insert "be") - page 5, first line: alpha should have two subscripts - page 5, six lines below Eq. (6): "there may also resonance" (a "be" seems to be missing in the sentence) - page 5, bottom: DFT is the standard acronym for "density functional theory". Using it for "discrete Fourier transform" as well, might cause confusion. - page 6, caption of Fig. 2: "logarithmically sampled" - page 7, 5 lines below Eq. (11): "As the frequencies of the external fields are muliples of ..." (no komma here) - page 7, 6 lines below Eq. (11): no "then" at the beginning of the line - page 10, bottom paragraph: The phrase "In Fig. 4, we report ..." is doubled. - page 12, line 4 of section 5.2: "Similar to what was observed ..." (insert "was") line 5: "... while the weak A and B excitons ..." (delete "the")

We thank the referee for the positive of our manuscript reporting these typos that we corrected in the new version of our manuscript

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

Answer to Referee 4

[...] I believe the manuscript is an important contribution in the field of nonlinear response of materials. The studied effects are relatively unknown, and the level of the developed theory is clearly cutting edge, as it includes many-body excitonic effects that play an important role in 2D materials like the ones studied in the manuscript. Therefore, I recommend the publication in SciPost Physics.

We thank the referee for their evaluation which recognize the significance of our work.

1- For clarity, it would be nice if the authors could choose a different letter from "P", or at least its calligraphy, to denote polarization and momentum, otherwise the derivations are hard to follow at times. This is specially true in sections like 3.1 (page 6); right after Eq. 9, the authors refer to "complex Fourier components $\vec{p}_n$", I assume that they want to refer to the quantity defined in Eq. 9 $\vec{p}^{(n)}$, but the symbol they employ actually corresponds to single-particle momentum introduced above Eq. 6.

To improve clarity, in the present version we have changed the calligraphy for the polarization and changed the Fourier coefficients $p^{(n)}$ to $C^{(n)}$ in the same manner as in Eq. (12).

2- The description on how the FI-SHG is computed at small finite frequency could be improved. According to Eq. 8, it appears that the response at +-2w is proportional to the third-order susceptibility at frequency set(\nu,\omega\omega). Then, I do not entirely understand the following sentence "summing the $\chi(3)$ extracted by P (2ω +) and P (2ω -)", one obtains the corresponding FI-SHG for low-frequency time-dependent pump fields." Do the authors mean that when \nu is positive (negative) it corresponds to 2w+ (2w-)? I believe this should be clarified.

A further minor point: the authors say "Among higher-order responses that can be extracted from Eq. (4), we look at the FI-SHG", but Eq. 4 explicitly discards higher-than-quadratic contributions. Could they clarify?

We now explicitly show higher order in Eq. 4 (now Eq. 6 in the new manuscript). In our procedure higher terms up to the machine precisions are extracted from the dynamics, though not explicitly used or plotted.

3- Why do the authors employ the approximate equality symbol in Eq. 5? To my understanding, this is the exact expression for the second-order susceptibility in perturbation theory.

The referee is right, we replaced the approximate equality with equality symbol.

4- The authors could provide more details on the symmetries of the two studied systems, e.g. their point group, and possibly provide an appropriate reference containing more details on the systems to help the interested reader.

We added a discussion and references on the symmetries of the two systems and their nonlinear response.

5- The authors rescale the response by a effective thickness d_eff. Is this procedure standard for quadratic responses? Do they expect any non-trivial dependence on this parameter?

For two-dimensional systems, the response is ill-defined. Then, to have a well-defined response function it is customary in experiments to assume an effective thickness, which is usually close to the layer-layer distance in the corresponding bulk system. In calculations, for two-dimensional systems we use a supercell approach and the response output by the calculation then depends on the supercell non-periodic dimension. In complete analogy with what is done in experiment (so making possible to compare with experimental results), instead of the arbitrary supercell non-periodic dimension, we use the effective thickness. The dependence on the thickness parameter is trivial, in the sense that is just a rescaling.