Answer to report 3

We would like to thank the referee for the positive review of the paper and for his/her questions and criticisms that helped us to improve the clarity of the present work.

In what follows we provide detailed answers to the questions raised by the referee. Modifications have been implemented in the main body of the text to address the points covered in this review.

My understanding is that the RWA has been used within the two-level system model. To my knowledge, this is not a good approximation when ultrashort pulses are used. Can the authors comment on that? Can that be the reason for the disagreement observed for the high field?

Yes, the RWA has been used in the analysis of the system at the TLS level. We have verified that this approximation does not introduce relevant errors in the present case. This check has been performed in two ways. First, the good agreement between TLS and *ab initio* simulations in the low field regime represents an indirect verification of the negligible impact of the RWA (since the *ab initio* simulations are without the RWA approximation). Second, as a further direct analysis, we have performed an extra *ab initio* simulation in the high field regime using only two bands (see Fig. 1). The results of this computation show no significant discrepancy with the with the corresponding TLS result, thus confirming the goodness of the RWA approximation for the results discussed in the present manuscript.

If the ab-initio approach is reduced to two bands, should one observe the same discrepancies with the converged simulation as for the TLS? did the author conduct such a test?

The number of bands in the *ab initio* simulations has been deeply analyzed. The results presented in the manuscript are built with 7 bands, 3 occupied and 4 empty bands (in this condition each k-point can give rise to one or several TLS depending depending on the local band energy structure). If we perform *ab initio* simulations with only 1 valence and 1 conduction bands, the results are almost identical to the one provided by the corresponding TLS (in this condition we have only 1 TLS per k-point). The comparison is shown in Fig. 1. This is a further confirmation that the discrepancies between the two approached are due to multi-bands effects that the TLS in not able to include.

Computational details related answers

Here we report the computational details related answers. These further information have been added in the main body of the text in section 2.

a) We have used a scalar relativistic norm conserving pseudopotential for the Ga and As atoms.

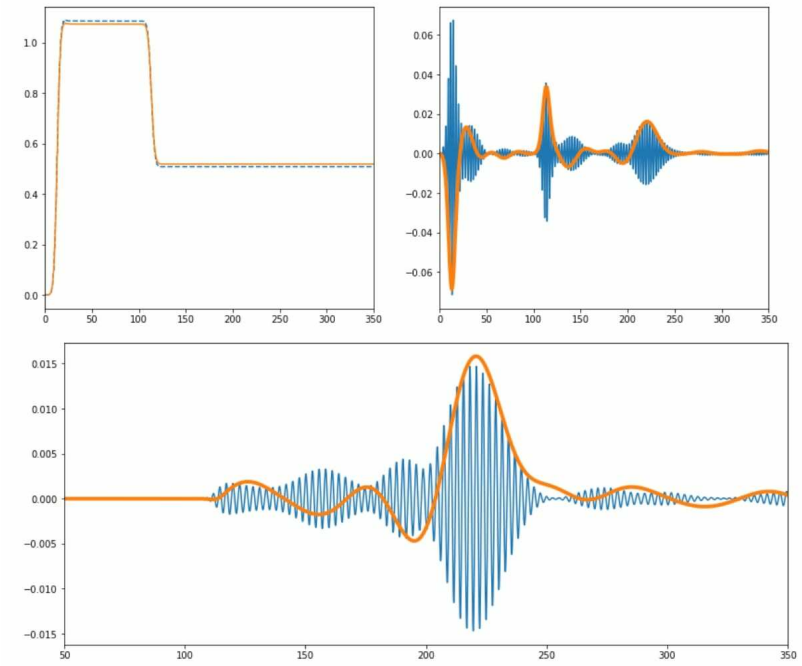

Figure 1: Comparison of the *ab initio* and TLS results using only a couple of bands. Top panel: (left) number of carriers, (right) polarization signal. Bottom panel: echo signal.

b) The units of a_{lat} have been added.

c) A rigid shift of 0.955 eV is used to open the DFT band gap.

d) Spin orbit coupling has not been included in the analysis. This is a reasonable approximation for the study of the observable (the electronic polarization) analyzed here. Indeed the position dipoles that are used to build the electronic polarization do not depend on the spin variables and, moreover, the inclusion of the SOC produces a splitting of the spin polarized bands of few meV, a values negligible w.r.t. the energy of the transitions activated by the pump (see also Fig. 1). Instead SOC becomes relevant in the analysis of the spin polarization, that we have presented in [1], and in that case a full relativistic pseudopotential has been used and the SOC has been included.

e) For what concerns the bands used in the computation see the answer to the previous point.

About the usage of the notation $P^{(i,j)}$

We have modified the expression for the cubic term as $P^{(3)}$ after equation 8, so that the notation $P^{(i,j)}$ is used only after its definition.

Lastly, the hyphenation pointed out by the referee have been corrected.

References

[1] M. D'Alessandro and D. Sangalli. Real-time modeling of optical orientation in gaas: Generation and decay of the degree of spin polarization. *Phys. Rev. B*, 102:104437, Sep 2020.

[2] W.-R. Hannes, A. Trautmann, M. Stein, F. Schäfer, M. Koch, and T. Meier. Strongly nonresonant four-wave mixing in semiconductors. *Phys. Rev. B*, 101:075203, Feb 2020.