Answer to report 4

We would like to thank the referee for the positive review of the paper.

Concerning the question around eq. 3, the dipole operator is computed from the *ab initio* (DFT) results. In particular it is represented in the Kohn-Sham basis, where its matrix elements are computed as

$$\langle n\mathbf{k}|\hat{\mathbf{r}}|m\mathbf{k}\rangle$$

Since the position operator is ill defined in periodic system, the previous formula is rewritten by expressing the commutator of $\hat{\mathbf{r}}$ with the Hamiltonian, that is

$$\langle n\mathbf{k}|\hat{\mathbf{r}}|m\mathbf{k}\rangle = \frac{\langle n\mathbf{k}|[\hat{H},\hat{\mathbf{r}}]|m\mathbf{k}\rangle}{(\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}})}.$$

The intra-band terms, i.e. the terms for which $(\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}}) < 0.01\ meV$, are neglected. The physics of intra-band transitions have been shown to be important, for photon-echo and four wave mixing experiments, when the laser pulse is off-resonance compared to the band to band transition energy. In the present case, where the laser pulse frequency is tuned resonant, the effect is expected to be negligible [2]. We also observe that this is equivalent to what is done in the TLS approach, where only dipoles between different states are included.

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.