Response to Referee #1

1. *The goal of the paper is to maximize observables that can described with Eq 6 by modifying field parameters using gradients. The computation of G(u) depends on the time trajectory, which could be also computed using the adjoint sensitivity method. I encourage the authors to include this in the discussion of their work.*

   We agree with the referee that discussing on the possible use of the adjoint method – or of other methods used by the quantum optimal control community – would improve the manuscript. In fact, we tried first using the adjoint method. It is, after all, the main method implemented in the code that we have developed and used for this and other projects (`https://qocttools.readthedocs.io/`). However, we could not make the adjoint method work in this case, and we are not sure that it can be applied, at least not with full generality.

   The reason is the following: In the adjoint method, in order to compute the gradient of the target function, one needs to solve for the *state* equation (in our case, Lindblad's equation), and for the *adjoint* equation. But in this context, the solution that is required in both cases is the periodic solution – in contrast to the usual case, in which one has an initial value problem. The existence of a periodic solution is not always guaranteed: While we could always find periodic solutions to the control equation, we found that the adjoint equation not always had a periodic solution.

   However, we are still not sure enough to make a statement such as "the adjoint method is not suitable for this problem", as there may be ways to use it that we have not been able to find, and which perhaps we wish to explore in the future. We have added some remarks about this to the manuscript (next-to-last paragraph in Section 2), as suggested by the referee.

2. - *As indicated by the authors the most similar paper is Ref. 28, where it was previously demonstrated the possibility to evaluate the jacobian of an ODE's steady-state with respect to the parameters that govern the dynamical evolution. The presented work expanded this idea to time periodic systems, however, there is a lack of notation and clarity.*
   - *Are Eqs. 15-17 similar to Eqs. 4-5 from Ref. 28? If so, the authors should indicate it and explain its differences.*
   - *I believe the following term is ambiguous, "Although to our knowledge, no previous work has attempted the optimization of NESSs with respect to the external drivings, a related work [28] ... ". Parameters that describe the system and control parameters globally affect the system so, under Ref 28's framework, one could also compute such gradients with respect to the control fields.*

   We combine the answer to these three points, as they refer to the same issue, i.e. the relationship of this work with Ref. [28].

   The term "non-equilibrium steady state" (NESS) may refer to different kinds of objects – and the objects that the authors of Ref. [28] address are different to the ones that we are addressing in this work.

   One kind of NESS may appear, for example, when a system, in the absence of external time-dependent perturbations, is in contact with two reservoirs at different temperatures. The steady state that may develop will be time-independent – but it will not be an equilibrium ensemble, such as the canonical ensemble. If Lindblad's equation is used to model that system and $\mathcal{L}$ is the (time-idependent) Lindbladian, $\rho_{\rm NESS}$ is a steady state if:

   $$\frac{\partial \rho_{\rm NESS}}{\partial t} = \mathcal{L}(\rho_{\rm NESS}) = 0\,.$$

   This is the situation that was addressed in Ref. [28], and the method developed there was based in the fulfillment of this equation. Thus, the function F in Eqs. (2), (4) and (5) in that reference is the time-independent Lindbladian (although we suppose that this line of reasoning can be extended to other master equations).

   The kind of NESS that we are addressing in this work is different: the Lindbladian is *time-dependent*, and therefore the NESS is not static; its time derivative is not zero: the condition that it fulfills is the periodicity, not the time-independence:

   $$\rho_{\rm NESS}(t+T) = \rho_{\rm NESS}(t)\,.$$

   In between t and $t+T$, the state changes with time, a change often referred to as *micromotion* (it can be argued that, in consequence the "steady state" name should not be used, but it has now become common usage).

   Therefore, Eq. (4) in Ref. [28] does not describe our case, and the fixed point theorem, (5) in Ref [28] does not apply here. The equation may seem similar to our Eqs. (15-17) inasmuch as they are linear equations, but there is an important difference: the operator $\overline{\mathcal{L}}$ in our equations is not a Lindbladian, but an *extended* Lindbladian: it is defined in the Floquet-Liouville space: the tensor product of the space of periodic square-integrable functions in time and the space of density matrices (or operators).

Note, however, that probably the method in Ref. [28] could be extended, by considering that F is not a time-independent Lindbladian, but a positive map linking the density matrix at time t with the evolved density matrix at time $t+T$. In this way, the fixed point equation could enforce the periodicity of the evolution of the density matrix. For example:

$$F(\rho(t)) = \rho(T) - \rho(t) = 0 \tag{1}$$

The resulting equations are probably much more involved.

We agree with the referee that our text does not make these differences clear enough. We have changed the text accordingly, at the end of the Introduction, and also in Section 2, the paragraph around current Eq. (20).

3. *Why was sequential Least-Squares Quadratic Programming algorithm the only used optimization proto- col? Briefly discuss if this method present an advantage over other more standard ones, e.g. BFGS or other.*

   The implementation of the SLSQSP algorithm in the nlopt library allows for the enforcements of bounds and constraints on the parameter search – a feature that is not allowed by the BFGS implementation in that library (this does not mean that the BFGS cannot be extended to be used with bounds, as there are other implementations that permit to use them). We do not think that the SLSQP is better than the BFGS algorithm for these problems in the absence of bounds – in fact we found quite similar results with all the gradient-based algorithms that we tried.

4. *I find the title misleading as engineering of quantum states could also be done with grid or sampling based approaches. The core result of the paper is an algorithm to compute gradients for the floquet steady-state, why not include that in the title of the paper?*

   We prefer short titles, and hence the rather generic title.

   However, since a more specific title is a request of two of the reports, we propose "Floquet engineering non-equilibrium steady states on the optimization of system properties with gradient-based methods"

5. *Increase the font size.*

   We cannot, at least using the journal's LaTeX class and template, which we assume is the recommended procedure for submissions.

6. *Regarding the normalization condition,* $\mathrm{Tr}\rho(u) = 1$*. For Lindblad-type systems, the Liouvillian is trace preserving, which indicate that* $\mathrm{Tr}\rho(u) = 1e$ *is an unnecessary constrain.*

   We do not need to add that condition to the numerical algorithms; as the referee points out it is fulfilled automatically by the Lindblad equation structure. However, we do need it to invoke it in the mathematical derivation leading to Eq. (19).

7. *Include a figure that shows the convergence of the search problem over the number of iterations. Additionally, are the field parameters initialize randomly?*

   Yes, we start the calculations from randomly generated initial guesses. We have added this information to the text (end of Section 3) and, as requested, added a plot with typical convergence histories for the optimization runs.

8. *The authors are missing some relevant citations:*

   - *Phys. Rev. Research 4, L012029 2022*
   - *arXiv:2011.12808*
   - *Lev Semenovich Pontryagin, EF Mishchenko, VG Boltyanskii, and RV Gamkrelidze. The mathematical theory of optimal processes. 1962.*
   - *The Implicit Function Theorem: History, Theory, and Applications (Berlin: Springer), S G Krantz and H R Parks (2012)*

   We have added those references as suggested by the referee.