Multi-objective genetic algorithm optimization of 2D- and 3D-Pareto fronts for vibrational quantum processes

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Abstract. A multi-objective genetic algorithm is applied to optimize picosecond laser fields, driving vibrational quantum processes. Our examples are state-to-state transitions and unitary transformations. The approach allows features of the shaped laser fields and of the excitation mechanisms to be controlled simultaneously with the quantum yield. Within the parameter range accessible to the experiment, we focus on short pulse durations and low pulse energies to optimize preferably robust laser fields. Multidimensional Pareto fronts for these conflicting objectives could be constructed. Comparison with previous work showed that the solutions from Pareto optimizations and from optimal control theory match very well.

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1. Introduction

The control of light–matter interaction is frequently performed with genetic algorithms (GA) in closed-loop experiments, where mask functions, consisting of a phase and a transmittance function, are optimized and used in a shaper to modulate the spectrum of an ultrashort, incident Fourier-limited (FL) laser pulse [1, 2]. In theory, either optimal control theory (OCT) [3]–[5] can be applied or in analogy to the experimental approach, the optimal laser pulses can be identified with the help of GAs and feedback signals, calculated with the knowledge of the molecular Hamiltonian [6]–[8]. As an advantage, the experimentally accessible parameter space, of the FL-pulses and the shaping technique can be scanned directly and consequently reliable predictions for the experimental search strategy and results can be made. The investigated control aims are fundamental state-to-state transitions as key elements of unitary transformations, implemented in the transition metal carbonyl W(CO)$_6$ and a CNOT operation for MnBr(CO)$_5$. Besides high quantum yields, we focus on additional features of the laser fields driving the quantum processes. We aim to reduce the pulse duration and the overtone excitation as far as possible, which is crucial for quantum control scenarios, where decoherence sets an upper limit to the laser–molecule interaction. Additionally, we are interested in the interdependency of these properties with the quantum yield. For such investigations multi-objective GA are well suited, allowing the control of several objectives at the same time. The optimal results form a solution space, known as the Pareto front.

2. Multi-objective GA and application

GA are the most commonly employed types of evolutionary algorithms. The approach finds a solution to a problem by using principles from genetic evolution, such as mutation, selection and crossover. For the first generation, a starting population of individuals is randomly generated, where each individual, i.e. each solution, is described by a decision vector $x = (x_1, x_2, \ldots, x_n)$ in the decision or parameter space $X$ of dimension $n$. The fitness of every individual is evaluated. Afterwards, a selection of the individuals is performed and they are randomly recombined and mutated to build a new generation, which is evolving to find better solutions for the control problem. In a single-objective algorithm the fitness is determined by assigning each solution to an objective value $y$ in the one-dimensional objective space $Y$ according to $f : X \rightarrow Y$. A solution $x^{(1)} \in X$ is better than another solution $x^{(2)} \in X$ if $y^{(1)} > y^{(2)}$. All solutions existing in the parameter space $X$ are mapped on $Y$ and a single optimal solution is the result of a single-objective GA run. In a previous study [9] with GA, where two conflicting goals should be accomplished simultaneously, we mapped both the criteria on to a single objective value $y$ or in other words, we included a cost function in the objective besides the original optimization aim [10]. This optimization strategy leads to a compromise of both control aims similar as in OCT but the interdependency of the two single objectives cannot be traced clearly. Multi-objective optimizations, also known as multi-criteria optimizations, are best suited for such multidimensional control tasks. Now, a solution is assigned to an objective vector $y = (y_1, y_2, \ldots, y_k)$ with the dimensionality $k$, given by the number of objectives [11]. The decision as to which solutions are better than others is more complex and is made with the selection operator. Here, the concept of the Pareto-dominance relation is applied. An objective vector $y^{(1)}$ prevails all other vectors $y^{(j)}$ if no component $y^{(1)}_i$ is smaller than the corresponding components $y^{(j)}_i$ and at least one component has to be larger. Such solutions are said to be non-dominated.
Infeasible

Feasible

point

Pareto point

Objective 1

Objective 2

Pareto front

Figure 1. Two-dimensional-(2D) Pareto front (red line and dark green circles). The light green dots are feasible points, whereas the yellow ones indicate infeasible solutions, with respect to the constraints.

and they can be mapped on different objective vectors. Consequently, a Pareto-optimal set of solutions is obtained, which build up the Pareto front (figure 1) in the objective space and the front represents the varying impact of the individual objectives. In this study, the Elitist non-dominated sorting genetic algorithm II (NSGA-II) [12] is applied, which has already been used in quantum control experiments [13]. Additionally, various supplementary constraints might be included in the NSGA-II algorithm. If a solution violates a constraint, it is an infeasible solution and discarded from the set. A schematic sketch of the Pareto front with feasible and infeasible solutions is depicted in figure 1. The multi-objective algorithm is applied to design optimal laser fields for molecular control scenarios, where it is desirable to control additional features on the outcome simultaneously. With this algorithm several control objectives can be maximized or minimized, such as the quantum yield, the pulse duration, the degree of overtone excitation, the complexity of the pulse and many more. The laser fields are constructed by theoretically sending a FL-pulse through a shaper and calculating the feedback signal with the knowledge of the Hamiltonian. The genome of the GA comprises the phase and transmittance function.

The bandwidth-limited pulses have a Gaussian shape and are centered at the carrier frequency $\omega_c$:

$$E(t) = E_0 \exp\{-\frac{1}{\tau_G}t\} \cos(\omega_c t) \quad \text{with} \quad \tau_G = \frac{\tau_p}{\sqrt{2 \ln 2}}.$$

The pulse duration $\tau_p$ is defined as the full-width at half-maximum (FWHM) of the intensity profile and the spectral width $\Delta\omega_p$ is the FWHM of the spectral intensity.

$$E(\omega) = E_0 \exp\left\{ -2 \ln 2 \left( \frac{\omega - \omega_c}{\Delta\omega_p} \right)^2 \right\}.$$

$E_0$ is the maximum intensity, $t$ the time and $\omega$ the frequency. The phase $\phi(\omega)$ is set to zero for bandwidth-limited pulses. After passing a shaper with the transmittance function $T(\omega)$ and the
The quantum control studies are based on vibrational excitation of metal carbonyls with ultrashort, phase and amplitude modulated laser pulses. As model system for selective vibrational excitation and for molecular quantum computing [17] we take the two transition metal complexes W(CO)$_6$ and MnBr(CO)$_5$, whose controllability with shaped pulses has already been studied [18, 19] to some extent. In particular, coherent control experiments on the T$_{1u}$ mode of W(CO)$_6$ have been successfully performed with the aim to maximize a state-to-state population transfer [20]. We briefly summarize our theoretical setup, further information can be found in [9] and references therein. The potential energy surfaces and dipole moment of the A$_1$- and E-symmetric stretching mode of MnBr(CO)$_5$ and one T$_{1u}$-symmetric mode of W(CO)$_6$ were calculated with the quantum chemistry package GAUSSIAN03 [21]. The quantum dynamical calculations were performed in the eigenstate representation. For the quantum gate calculations, we define the vibrational ground state as the qubit state $|0\rangle$ and the first vibrational excited state as $|1\rangle$.

### 3.1. 2D-Pareto fronts for state-to-state transitions

Possible routes for highly efficient state-to-state population transfer are investigated theoretically for a T$_{1u}$-mode of W(CO)$_6$. The fundamental transition $0 \rightarrow 1$ participates as one key element in the corresponding molecular unitary transformations. The dependence of the quantum yield on different factors, referring to the pulse properties and the control mechanisms
Table 1. FL-pulse properties for state-to-state transitions.

| Parameter | Min  | Max  |
|-----------|------|------|
| $a$       | 0    | 10   |
| $b$       | 20 000 | 60 000 |
| $E_0$     | 0.00001 au | 0.001 au |
| $\tau_p$  | 100 fs | 2000 fs |
| $\omega_c$ | 1950 cm$^{-1}$ | 2020 cm$^{-1}$ |

is explored. Simultaneously to the maximization of the quantum yield, we aim for minimal temporal pulse duration, pulse energy and intermediate overtone population. These additional features are relevant, when decoherence sets an upper limit for the laser–molecule interaction and dissipative effects will be most disturbing, when intermediate overtone excitation is high. Within the experimentally available range, we minimize the pulse energy to prevent side effects, such as nonlinear processes. We will use the experimentally investigated molecule W(CO)$_6$ as candidate to study different Pareto fronts. The presented multi-objective GA (MOGA) calculations are performed with a population size of 60 and 500 generations.

3.1.1. Objectives: quantum yield and pulse duration. The MOGA offers an elegant possibility to study the relation of the quantum yield and the pulse duration of shaped laser fields, which corresponds to the effective laser–molecule-interaction time. From our previous GA study [9], we know that the quality of the solutions depends on the FL-pulse properties. Therefore, we extend the genome of the GA, which comprises in the experiment only the mask parameters, by the FL-pulse properties, the FWHM $\tau_p$, the carrier frequency $\omega_c$ and the maximum energy $E_0$. We perform Pareto optimizations for analytic and pixeled mask functions. As a first test, we use pixeled mask functions with a spectral pixel resolution of 10 cm$^{-1}$ for all calculations presented in section 3.1. For the incident pulse, we use the parameters given in table 1. As an alternative, an analytic form $\phi(\omega) = a \sin(b\omega)$ for the phase function is applied as often used in the experiment [14]–[16] and especially in the mid-IR regime [20] and we optimize both phase parameters $a$ and $b$. In total, five parameters have to be adjusted by the GA, given in table 1 for analytic phase functions. If pixeled mask functions are applied with 20 pixels for each function (phase and transmittance), 43 parameters including the FL-pulse properties are optimized.

The Pareto fronts for the maximization of the quantum yield and minimization of the pulse duration are depicted in figure 2. We define the duration of a shaped laser field as the period of time, during which the field $|E(t)|$ exceeds a threshold value of $5 \times 10^{-6}$ GV cm$^{-1}$. Below this energy the mid-IR laser interaction with the carbonyl complexes is negligible. In the upper panel, pixeled mask functions and in the lower panel analytic phase functions are optimized. The efficiencies are shown from 25%–100% and the high-fidelity region is zoomed in, in the smaller insets. Both Pareto fronts are convex with respect to the feasible solutions, which is the simplest case for MOGA optimizations. The convex behavior implies that a high quantum yield and short pulse durations are objectives which are reconcilable to some extent. The fronts are characterized by two branches, the vertical one corresponds to high quantum yields and the horizontal one to short pulse durations. A main difference, traced from figure 2(a) and (b), is that in the case of an analytic phase modulation the durations of the pulses, reaching high
efficiencies, are clearly longer. The FWHMs of the initial FL-pulses are in the same range for both approaches. They are found to be rather long (1.0–1.5 ps) and the analytic, sinusoidal phase modulation cannot generate multipulses for the solutions on the Pareto front in figure 2(b). Thus, the long durations of the sinusoidal phase modulated pulses are not due to large subpulse shifts in the multipulse, but rather caused by the unflexible sinusoidal phase approach.

In figure 3, the best solutions (>99%) for the pixeled mask functions and the analytic phase are depicted together with the corresponding, scaled spectra and their mask functions. The pixeled phase function (left) is rather flat and the shaped pulse provides a duration of ~8 ps. In contrast, the sinusoidal phase (right) leads to a temporal duration of ~20 ps. We monitored the efficiencies of all laser fields exceeding 80% (figure 4, top). For the analytic phase (black dots), we find 30 solutions and 22 for the pixeled functions on the Pareto fronts. Additionally, the FL-pulse properties are given in the lower three panels. There is no explicit trend for these properties, instead, in the high-fidelity region we find different solutions with optimal parameters in a broad range, i.e. the FWHMs reach from 900–1880 fs, the maximum energy from 0.001–0.0033 GV cm\(^{-1}\) and the carrier frequency from 1984–1991 cm\(^{-1}\). This very
Figure 3. Resulting best laser fields for the two objectives: high quantum yield and low pulse duration, in the upper panels. The respective mask functions (red: phase; blue, dashed: transmittance) and scaled FL-pulse spectra (solid, black) are depicted in the lower panels. Left: pixeled mask functions. The solid black line refers to the spectrum of the FL-pulse, whereas the black dashed line shows the shaped, scaled spectrum. Right: analytic phase modulation.

Figure 4. For the high-fidelity region of the Pareto fronts from figure 2, the yield and the FL-pulse properties are depicted. The black dots refer to an analytic phase modulation and the red triangles to pixeled mask functions.
Figure 5. Resulting Pareto fronts for the two objectives: high quantum yield and short pulse duration, where the phase range is confined, i.e. scaled by the factors $f$ given in the legend.

Advantageous result means that the subspace of different control parameters is rather flexible and optimal solution, matching best with the experimental conditions, can be selected from the set. A very obvious distinction can be found in the magnitude of the maximum energy, which is always larger for pixeled mask functions, where additional amplitude modulation is possible. This can be seen in figure 3, left side, where a pronounced spectral damping by the transmittance function is induced. Accompanied by the fact that the carrier frequencies for pixeled mask functions are detuned by $5 \text{ cm}^{-1}$ (figure 4, bottom) compared with the analytic functions, the transmittance suppresses the major spectral component (figure 3, left side), to reach a higher frequency resolution.

From our previous work [9], we know that the complexity of the envelope function, in case of the pixeled mask functions, can easily be reduced by decreasing the co-domain of the phase function by a factor $f$ (given in the legend) to the range $[0, 2\pi] \cdot f$. The effect of limited phase ranges is shown in figure 5. We find that smaller ranges benefit the MOGA search. In general, phase variations between each pixel are much more probable than flat phase functions, but lead to longer pulse durations and complex structures [9]. To find optimal solutions with respect to both objectives, either the number of GA generations can be enlarged significantly or as demonstrated in figure 5 the co-domain of the phase can be confined.

Only a small part of the complete Pareto front (figure 2), the high-fidelity region, exceeding efficiencies of 99.0% is relevant in the context of quantum information. If such a constraint is introduced and not fulfilled by a solution, this infeasible point is discarded. Consequently, the algorithm searches more intensively in the region of the feasible solutions, which become predominantly located in the high-fidelity regime. This region is better sampled now, since we use the same number of generations and population as before. The fidelity is limited to a minimum of 99.0% and a corresponding calculation for the pixeled mask functions leads to a Pareto front, more dense in the regime from 100–99% (figure 6). The black symbols indicate the position of all feasible solutions in the objective space and the red dots of the Pareto-optimal solutions. The front is steeper and dominates the previous front without constraint (shown in figure 2(a) and for comparison indicated in figure 6 by the light blue dots). We found significantly shorter FWHMs of the FL-pulses in the range of 590 fs, which is, however,
a statistical finding. This can be traced from several MOGA runs with efficiency-constraints, but with random starting individuals. In each run, we found high-fidelity Pareto fronts, where the solutions converged to different FL-pulse FWHMs, distributed in the complete range used (table 1). All these solutions must also be found in the unconstrained search when the number of iterations is enlarged. We can conclude that in case of pixeled mask functions a very broad FWHM range at least from 100–2000 fs, appropriately combined with the remaining FL-pulse parameters, can lead to highly efficient, short laser pulses for state-to-state transitions.

3.1.2. Minimization of the pulse energy. The pulse energy, a further crucial feature, was unconfined so far and no bias on the energy-efficiency of the transfer process was implemented. Analogously to OCT, where a penalty energy function is included, we incorporate the minimization of the energy of the shaped field ($\int_0^T E(t)^2 \, dt$) as the second objective besides the maximization of the quantum yield. The resulting Pareto front is depicted in figure 7(a), for pixeled mask functions. No constraint on the phase range is used and the FL-pulse parameter ranges are given in table 1. The minimum pulse energy of the Pareto-optimal solutions has to exceed $3.4 \times 10^{-4} \text{J cm}^{-2}$ for an efficiency of 99.0%. There are several optimal points on the front, exceeding efficiencies of 99% with similar envelope functions for the modulated laser fields. The rather short pulse durations in the range of ~8 ps, which we obtained before, when the minimization of the pulse duration was the second control aim cannot be maintained. The temporal duration of the best pulse now exceeds 20 ps (figure 7(b)). The best pulse is obtained with a transmittance function strongly damping the spectral signal of the FL-pulse. This is one possible solution, also often observed in OCT. Less energy waste can be obtained when the FL-pulse energy is limited. To reach all desired features at the same time, the three objectives, low energy, short pulse duration and high quantum yield, have to be defined. Corresponding results will be presented in section 3.1 for a unitary transformation.
3.1.3. Minimization of the overtone population. A high degree of intermediate overtone population may be obstructive for quantum control experiments. Population relaxation times ($T_1$) in the condensed phase for carbonyl complexes are only half as long as the $T_1$ timescale of the first excited vibrational state or even shorter [20]. A low overtone excitation can be reached indirectly by using long FL-pulses, but the amount of overtone population can also explicitly be formulated as a second objective. We minimize the population $P_i(t)$ of all $i$ overtone states, i.e. $\sum_{i=2}^{N} \int_{0}^{T} P_i(t) \, dt$, where the time is used in atomic units. The corresponding Pareto front is shown in figure 8(a). For our setup, we find that still a finite amount of overtone excitation has to be taken into account for highly efficient solutions. A value of 0.05 integrated population corresponding to an intermediate population maximum of 16% in the second vibrational state is still necessary. Solutions with confined phase ranges (figure 8(b)) cannot compete with the calculation for the complete range $[0, 2\pi]$ and we find that a flexible phase functions turns out to be essential here. A progression of the Pareto front similar to the pixeled phase (figure 8(a), red symbols) is obtained when a sinusoidal phase modulation is applied on the rather long FL-pulses (blue symbols). Consequently, these analytic phase functions allow to suppress the amount of population transfer in higher vibrational levels equivalently.
Figure 8. (a) Result of the MOGA optimization with the two objectives high quantum yield and low overtone population. Black and red symbols: for pixeled mask functions. Blue symbols: for analytic phase functions. (b) Pareto fronts, with confined co-domains \([0, 2\pi \cdot f]\) of the pixeled phase function, where the factors \(f\) are given in the legend.

The analysis of the 2D-Pareto fronts allowed to understand and interpret the interplay of the quantum yield with additional features of interest for the first time, qualitatively as well as quantitatively. From the 2D fronts it is also possible to judge whether higher dimensional fronts are needed to improve the solutions with respect to their feasibility.

3.2. 3D-Pareto front for a unitary transformation

All objectives presented in section 3.1 are desirable to be reached simultaneously for effective, robust and realizable quantum gate operations. We discuss the effect of the three most relevant objectives: the maximization of the quantum yield, the minimization of the shaped pulse duration and the energy density per area. The quantum yield is defined as the averaged quantum efficiencies of the single population transfer processes induced by the quantum gate operation. To obtain a phase correct quantum gate, an additional time delay has to be attached, for correct phase rotation [22]. As an example, we take the two-qubit CNOT gate, operating on the E-mode of MnBr(CO)$_5$. The calculations are performed for pixeled mask functions with a
Figure 9. (a) 3D-Pareto front for the unitary transformation CNOT\(_E\) with the three objectives quantum yield, pulse duration and pulse energy. (b) CNOT\(_E\) gate, one of the solutions from the optimal set. The position is indicated on the 3D-Pareto front by the black dot.

With a spectral resolution of 10 cm\(^{-1}\) for 1000 generations and a population size of 100 individuals. Additionally, we applied a constraint of minimum 90.0% yield. The 3D Pareto front, shown in figure 9(a), is tilted in the 3D objective space and for better visualization it is interpolated. The highest efficiencies are shown as the red section and mark the region of unitary transformations that can be realized with minimal pulse energy and duration. For short pulse durations the section is slightly curved, while for increasing pulse durations the energy stays constant. For shorter pulse durations < 9 ps, intermediate overtone excitation occurs and high energies are necessary to depopulate these states again. For longer pulse durations the spectral resolution suffices to avoid overtone excitation and the required energy only depends on the size of the fundamental transition matrix element. The Pareto front tilts towards lower quantum yields for smaller pulse energies and durations. The pulses, found as Pareto-optimal solutions provide very simple envelope functions. Most of them are almost Gaussian-shaped or composed by Gaussian pulses overlapping in time. The quantum gate laser field with the highest efficiency is presented in figure 9(b). The pulse energy is 11.8 \(\times 10^{-4}\) J cm\(^{-2}\) and the pulse duration is 12 ps. The Pareto-optimal pulse shapes are very similar to our previous OCT results [18, 23]. Here, the duration was varied manually to obtain simple pulse structures, in the MOGA calculation, the pulse duration was selected as an objective. Now, the solutions found by the search strategies OCT and MOGA clearly coincide, which means that the MOGA results match the OCT subspace or vice versa the simple structured OCT results are located on the Pareto front.

Different co-domains \([0, 2\pi] \cdot f\) for the phase functions are compared in figure 10, where a spectral pixel resolution of 10 cm\(^{-1}\), a population size of 60 and 500 generations were used. In figure 10(a) the 3D-Pareto fronts for the factors \(f = 1.0, 0.5, 0.1\) and 0.0 are depicted, figures 10(b)–(d) show projections onto two of the objectives. From figure 10(b), we can trace the relation of the pulse energy and the pulse duration. The pulse durations become shorter for confined co-domains, e.g. for pure amplitude modulation (\(f = 0\), cyan dots) the mean pulse duration is \(\sim 7\) ps compared with \(\sim 14\) ps for full phase modulation (red dots), which can also be seen in figure 10(d). On the other hand, flexible phase functions lead to shaped pulses with the least pulse energy (red dots in figure 10(c)) and vice versa, the most energy is needed for pulses, which are amplitude modulated only. All 3D-Pareto fronts obtained for the factors \(f\) confining the co-domains would merge into one for a large number of generations. If one feature, such as...
the pulse duration, is of special interest the search can be accelerated significantly by reducing the phase range. If more, conflicting features are of interest, an appropriate compromise must be found, e.g. low pulse energy is facilitated by flexible phase functions, whereas low pulse durations are realized by small phase variations.

Changing the spectral resolution for the pixels from 10–5 cm$^{-1}$ allows the algorithm to converge faster, but leads to more complex envelope functions. Additionally, the pulse duration regime for high-fidelity solutions shifts from 7–15 to 8–27 ps. For experimental applications, we suggest using only the minimum necessary spectral resolution of the pixels so that the desired quantum yield can still be reached. This will lead to envelope functions with the lowest complexity, the shortest pulse durations and lowest energies, which are the robust solutions in the search space.

4. Conclusion

In this study, we used a multi-objective genetic algorithm to optimize mask functions for laser fields driving different quantum processes such as state-to-state transitions and unitary transformations. By default, a single-objective GA is used in closed-loop experiments, where the main emphasis is put on the quantum yield and no bias concerning e.g. the pulse duration
is included. Here, we theoretically optimize different features of the shaped laser pulse and the excitation mechanism simultaneously. Especially, we take into account the pulse duration, the energy density per unit area and the induced degree of intermediate overtone excitation. The Pareto fronts for each set of objectives are constructed. All of them are convex with respect to the dominated feasible solutions and consequently the selected objectives (quantum yield, pulse energy, pulse duration, intermediate overtone population) can be reconciled. The minimum requirements on the laser pulses or crucial features of the excitation mechanisms can be extracted from these fronts. We find that the highly efficient Pareto-optimal solutions can be realized with a large variety of FL-pulse parameters, which are very advantageous as it makes the subspace of control parameters rather flexible and an optimal solution, matching best with the experimental conditions, can be located and selected from the Pareto front. The analysis of the 2D- and 3D-Pareto fronts allowed to trace the interplay of various objectives of interest, qualitatively as well as quantitatively. For experimental applications, we suggest using only the minimum neccessary spectral resolution of the pixels and adapting the phase co-domain to generate short pulse durations. These results are assumed to be the most robust ones also against decoherence and as a positive side effect, they show very simple envelope functions. In addition, the MOGA and OCT predictions for the unitary transformations match very well. Their careful handling allows to find the simplest and most promising solutions for given control scenarios and reveals the experimental route towards them.

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