The Application of Evolutionary Multi-Criteria Optimization to Dynamic Molecular Alignment

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Abstract—This study introduces the multi-criteria approach to the optimization of dynamic molecular alignment by shaped femtosecond laser pulses, which has been considered so far only as a single-criterion problem. The paper applies advanced Pareto front approximation algorithms to this challenging real-world, high-dimensional, and computationally expensive problem, working with low-dimensional parameterizations of the electric field. Standard approaches (NSGA-II) and their metamodel-assisted extensions based on Kriging, are applied to this optimization task and compared among each other. The study confirms the conflicting nature of the objectives. Interesting features of the problem domain, such as the geometry of the Pareto front are revealed. Furthermore, metamodel-assistance, in particular pre-screening with the Kriging-based expected improvement criterion, proves to be a valuable ingredient for improving the numerical results.

I. INTRODUCTION

Recent technological developments have made lasers with pulse lengths on the order of femtoseconds routinely available, which allow to control the motion of atoms or molecules by irradiating them with laser light. In our research we focus on the alignment of molecules by a laser pulse, and in particular on the optimization of the pulse shape for the purpose of high alignment. From the optimization perspective, the task is the calibration of a one-dimensional phase function, which plays the key role in shaping the pulse. In this study we apply for the first time a multi-criteria approach to this so-called Femtosecond Laser Pulse Shaping problem. Function evaluations for this problem are time consuming and a single function call takes 35sec on a Pentium-4 of 2.6 GHz. Therefore, besides standard methods for evolutionary multi-criteria optimization (NSGA-II), we use metamodelling, i.e. fast surrogate models of the objective function gained by interpolation, as a means to accelerate the search. In this paper, instead of proposing a new approach, we adopt metamodel-assisted optimization algorithms that were introduced in [1], where they have been partly only applied on artificial test problems.

The remainder of the paper is organized as follows. Section 2 presents briefly the physics problem, namely Femtosecond Laser Pulse Shaping. In section 3 we introduce the multi-criteria optimization methods in use. This is followed in section 4 by the description of the experimental setup and the numerical results. In section 5 we draw conclusions, summarize our study, and propose future directions for it.

II. THE PROBLEM: LASER PULSE SHAPING

We provide here a short introduction to the given physics problem, limited to the scope and the framework of this paper. For more details of the physics problem, we refer the reader to [2].

A. General

The advent of modern laser pulse shaping techniques in the femtosecond regime has made it possible to control the motion of nuclei and even electrons by a judicious choice of the pulses shapes. The application to dynamic molecular alignment [3] is of considerable interest in this context because of its many practical consequences: a multitude of chemical and physical processes ranging from bimolecular reactions [4] to high harmonic generation [5] are influenced by the angular distribution of the molecular sample. Furthermore, in many fundamental molecular dissociation or ionization experiments the interpretation of the collected data becomes much easier when the molecules are known to be aligned with respect to a certain axis. Hence, techniques to generate molecular alignment are much needed.

The goal of our research is thus to optimize the alignment of an ensemble of molecules after the interaction with a shaped laser pulse. There is currently a great interest in the atomic and molecular physics community to align molecules with laser pulses, since dealing with an aligned sample of molecules simplifies the interpretation of experimental data. By applying a self-learning loop using an evolutionary mechanism, the interaction between the system under study and the laser field can be steered, and optimal pulse shapes for a given optimization target can be found. In our work, the role of the experimental feedback in the self-learning loop is played by a numerical simulation [6].

B. Numerical Modeling

To calculate the time-dependent alignment, the Schrödinger’s equation for the angular degrees of freedom of a model diatomic molecule under the influence of the shaped laser field is solved. Explicitly, the time-dependent profile of the pulse, which completely determines the dynamics after the transition to the rotating frame has been performed, is described by:

\[ E(t) = \int_{-\infty}^{\infty} A(\omega) \exp(i\phi(\omega)) \exp(i\omega t) \, d\omega, \]  

(1)
where \( A(\omega) \) is a Gaussian window function describing the contribution of different frequencies to the pulse and \( \phi(\omega) \), the phase function, equips these frequencies, which are equally distributed across the spectrum of the pulse, with different complex phases. Hence, by changing \( \phi(\omega) \), the temporal structure of \( E(t) \) can be altered. In a real life, physical, pulse shaping experiment \( A(\omega) \) is fixed, and \( \phi(\omega) \) is used to control the shape of the pulses. We have used the same approach in our numerical simulations, i.e. the search space is in the frequency domain while the fitness evaluation is performed in the time domain. To this end, we interpolated \( \phi(\omega) \) at \( N \) frequencies \( \{\omega_i\}_{i=1}^N \); the \( N \) values \( \phi(\omega_i) \) are our decision parameters to be optimized. In order to achieve a good trade-off between high resolution and optimization efficiency, the value of \( N = 80 \) turned out to be a good compromise.

Primary: Angle of Molecular Axis

The alignment’s quantity, i.e. the success-rate or fitness, is defined as the expectation value of the cosine-squared of the angle of the molecular axis with respect to the laser polarization axis. Hence, the fitness function assigned to a pulse shape, which is the primary objective, is given by:

\[
 f_1 = \max_{E(t)} \langle \cos^2 (\theta) \rangle
\]

subject to maximization. Shaped femtosecond laser pulses that lead to a high degree of alignment manage to maximize the number of rotational states that are in phase at these times. However, they have to fulfill an additional requirement: the description of the molecule as a rigid rotator is strictly valid only for low field intensities. The higher the applied intensity is, the more important other competing channels like dissociation and ionization become. Therefore, one would like to achieve good alignment while keeping the peak laser intensity as low as possible. For this reason recent publications have focused on finding pulse shapes other than a Fourier transform limited (FTL) pulse, a pulse given by setting a zero phase function, \( \phi(\omega) = 0 \). Its intensity, denoted by \( I_{\text{FTL}} \), is given by:

\[
 I_{\text{FTL}} = \max_t \left\{ \left( \int_{-\infty}^{\infty} A(\omega) \exp(i\omega t) \, d\omega \right)^2 \right\}
\]

The FTL pulse obtains a high degree of alignment. However, Leibscher et. al. [7], [8] have theoretically shown that in the nonperturbing regime a train of pulses lead to better alignment than a single FTL pulse.

Thus, since a high degree of alignment with a peak intensity as low as possible was the desired result, the second objective is the minimization of the pulse intensity. Previously, this goal has been tackled using a punishment term in a single-criterion optimization (see, e.g., [9]):

\[
 I_p = \int_0^T E^2(t) \cdot \Theta (E^2(t) - I_{\text{thr}}) \, dt
\]

where \( \Theta(x) \) is the Heaviside step function, \( I_{\text{thr}} \) was a parametric threshold, and \( T \) is some large time constant (here \( T = \infty \)). Thus, the previous single-criterion fitness function assigned to a pulse shape was:

\[
 F_{OLD} = \max_{E(t)} \left( \cos^2 (\theta) \right) - \lambda I_p
\]

By choosing \( \lambda \) large enough, it was shown that \( I_{\text{thr}} \) could be used to effectively operate the evolutionary algorithms only on a subset of pulses whose maximum peak intensity approached the threshold intensity from below [9].

Secondary: The Second Harmonic Generation

A laser pulse going through crystals produces light at the octave of its frequency spectrum. The total energy of the radiated light is proportional to the integrated squared intensity of the primary pulse. The time-dependent profile of the laser field is exactly as given in Eq. 1. The Second Harmonic Generation signal is then defined by:

\[
 SHG = \int_0^T E(t)^4 \, dt
\]

Second Harmonic Generation is a process that turns out to be a good test case in the physics lab, and its investigation contributes to the understanding of the alignment problem, due to the fact that the SHG is a measure of the spikiness of a pulse, and this property is useful for the definition of a punishment function. Thus, we choose to define it as the second objective, subject to minimization:

\[
 f_2 = SHG
\]

Note, however, that one can show analytically that the FTL pulse, i.e., a zero phase function, maximizes the SHG,

\[
 \arg \max_{\phi(\omega)} \{ \text{SHG}(\phi(\omega)) \} = 0.
\]

C. Related Work

This laser pulse shaping problem, based on numerical simulations, has already been tackled at several levels. A survey of modern evolutionary approaches was introduced [9], a new parameterization method was developed and investigated [10], and niching methods were applied to this problem [11].

III. Multi-Criteria Optimization

We apply multi-criteria optimization to the problem, and in particular we aim to find the Pareto front for the bi-criteria problem:

\[
 f_1(\vec{x}) \to \max, \quad f_2(\vec{x}) \to \min, \quad \vec{x} \in X = [x_{min}, x_{max}] \subset \mathbb{R}^{80}
\]

Here \( f_1 \) and \( f_2 \) denote the two objective functions. Furthermore, \( \vec{x} \) denotes the decision vector, taken from the multi-dimensional decision space \( X \). \( X \) is the hyper-box bounded by \( \vec{x}_{min} \) and \( \vec{x}_{max} \). Furthermore, we assume both objective functions to be independent, and to be black-box functions.

In Pareto Optimization one is looking for the efficient set for the multi-criteria optimization problem, and its Pareto front. For the readers who are not familiar with these concepts, and in order to clarify notation used in this paper, we
shall briefly review these concepts formally. Despite the application problem’s nature, and without loss of generality, we will for now assume that all objectives are to be minimized, noting that maximization can be easily reformulated as minimization by inverting the sign of the objective function.

A partial order is defined on the solution space \( \mathcal{Y} = f(\mathcal{X}) \) by means of the Pareto domination concept for vectors in \( \mathbb{R}^m \): For any \( \vec{y}_1 \in \mathbb{R}^m \) and \( \vec{y}_2 \in \mathbb{R}^m \): \( \vec{y}_1 \) strictly (Pareto) dominates \( \vec{y}_2 \) (in symbols \( \vec{y}_1 \prec \vec{y}_2 \)) if and only if: \( \forall i \in \{1, \ldots, m\} : y_i^{(1)} \leq y_i^{(2)} \) and \( \exists i \in \{1, \ldots, m\} : y_i^{(1)} < y_i^{(2)} \). Note, that in the bi-criteria case this definition reduces to: \( \vec{y}_1 \prec \vec{y}_2 \iff y_1^{(1)} < y_1^{(2)} \land y_2^{(1)} \leq y_2^{(2)} \lor y_1^{(2)} \leq y_1^{(1)} \land y_2^{(1)} < y_2^{(2)} \). In addition to the strict domination \( \prec \) we define further comparison operators: \( \vec{y}_1 \preceq \vec{y}_2 \iff \vec{y}_1 \prec \vec{y}_2 \lor \vec{y}_1 = \vec{y}_2 \). Moreover, we say \( \vec{y}_1 \) is incomparable to \( \vec{y}_2 \) (in symbols: \( \vec{y}_1 \nparallel \vec{y}_2 \) ), if and only if \( \vec{y}_1 \nprec \vec{y}_2 \land \vec{y}_1 \npreceq \vec{y}_2 \). For any compact subset of \( \mathbb{R}^m \), say \( \mathcal{Y} \), there exists a non-empty set of minimal elements w.r.t. the partial order \( \preceq \) (cf. [Ehr05, page 29]. Points of this Partial order are called non-dominated points. Formally, we can define a non-dominated set via: \( \mathcal{Y}_N = \{ \vec{y} \in \mathcal{Y} \mid \nexists \vec{y}' \in \mathcal{Y} : \vec{y}' \prec \vec{y} \} \). Having defined the non-dominated set and the concept of Pareto domination for general sets of vectors in \( \mathbb{R}^m \), we can now relate it to the optimization task: The aim of Pareto optimization is to find the non-dominated set for \( \mathcal{Y} = f(\mathcal{X}) \) and its pre-image in \( \mathcal{X} \), the so-called efficient set. In many practical applications we are also satisfied with a set of solutions the image of which (under \( f \)) yields a good approximation to the non-dominated set, though a definition of what is a good approximation is problem dependent. Often, it is desired to achieve a uniform distribution on the Pareto front and a good convergence of all points in the approximation set to some non-dominated solution.

A. Choice of methods

In order to select an appropriate optimization method, the following characteristics of the objective functions in the application problem are of importance: Based on our experiences with the application problem in the single objective domain, we assume that the functions \( f_1 \) and \( f_2 \) are continuous in most points, highly nonlinear and probable highly multi-modal. Nothing is known yet about the shape of the Pareto front for the application problem. Analytical techniques and methods based on differential calculus are likely to fail in this problem, because of the complexity of the integral equations [2].

Due to their robustness and flexibility, evolutionary multi-criteria optimization algorithms (EMOA) have recently received increased attention as problem solvers for such difficult simulator-based optimization problems [12], [13], [14]. Among these methods, the NSGA-II method is one of the most popular, and it has been successfully applied to many real-world problems.

B. NSGA-II

The NSGA-II algorithm has been proposed by Deb [12]. It aims to obtain an \textit{well distributed approximation set} of points that are close to the Pareto front. Both, closeness and diversity, are addressed in the selection operator, where the population is sorted lexicographically using non-domination ranks as primary sorting criterion, and crowding-distances as secondary sorting criterion. In nondominated-sorting, first the non-dominated set is identified and its members get assigned rank 0. Then, the non-dominated set from the remaining individuals is computed, and assigned rank 1, and so on, until all individuals are ranked (Fig. 1). To determine the crowding distance, a measure of diversity contribution, the circumference of the box touching the nearest neighboring solutions is computed (Fig. 2). Basically, the NSGA-II is an elitist EA with \((\mu + \lambda)\) selection, as outlined in algorithm 1. The \((\mu + \lambda)\)-EA starts with initializing the \textit{generation} counter \( t \). After generating the initial population with \( \mu \) individuals by uniform random sampling in \( S \), the EA generates a set

\begin{algorithm}
1: \( t \leftarrow 0 \)
2: \( P_t \leftarrow \text{init()} \) \{ \( P_t \in S^\mu \): Set of solutions \}
3: evaluate(\( P_t \))
4: \textbf{while} \( t < t_{\text{max}} \) \textbf{do}
5: \( G_t \leftarrow \text{generate}(P_t) \) \{Generate \( \lambda \) variations\}
6: evaluate(\( G_t \))
7: \( P_{t+1} \leftarrow \text{select}(G_t \cup P_t) \) \{Rank and select \( \mu \) best\}
8: \( t \leftarrow t + 1 \)
9: \textbf{end while}
\end{algorithm}
"G_t of λ new solutions through recombination and mutation. The new candidate solutions are evaluated and ranked in terms of their quality, i.e., the rank in the aforementioned lexicographical order. The μ best solutions in \( G_t \cup P_t \) are selected to form the new parent population \( P_{t+1} \). For a detailed description of the variation operators we refer the reader to Deb [12]."
The integral is computed numerically.

In this paper, the advanced EMOA have been applied for the first time on molecular alignment problems, and we will now give a detailed presentation of results. Note, that tests of the metamodel-assisted EA on artificial test problems have been performed in [1].

IV. EXPERIMENTAL PROCEDURE

A. Preliminary

In this study we are using a new numerical variant of the alignment simulation, which has not been optimized before in the single-criterion framework (for details see below). To assess the performance of our multi-criteria optimization on this variant, we have first conducted a set of single-criterion runs, based on the single fitness defined in Eq. 5. We have used $\lambda = 1$, and $I_{thr}$ was $0.36 \cdot I_{FTL}$ (physics parameters: $\Omega_{X A} = 1.8 \cdot 10^{14}$ s$^{-1}$, $T_{FWHM} = 100$ fs and the sample temperature was 100 K). Given the set of algorithms used in [9], 5 independent runs for each algorithm were applied with the goal of maximizing the alignment. Fig. 5 shows the evolution of the fitness for the best case of each of the seven different algorithms. There are two observations to be made from this figure: first, the standard genetic algorithm performs significantly worse than all the other methods, both in terms of convergence speed and the highest fitness value that was found. Second, most of the other algorithms perform more or less equally well, with the DR2 algorithm finding the “best optimum”. While the DR3 algorithm shows the fastest initial fitness increase, the slope of the fitness decreases already after about 10000 evaluations. We have found this behavior to be typical for the DR3 algorithm. In all the runs, the best solution obtained was directly the $\langle \cos^2(\theta) \rangle$ pulse was able to produce, i.e. pure alignment, without punishment. Thus, the highest alignment known to us in the current settings is $F_{OLD}^{GA} = 0.689$, whereas the best value obtained by the GA was $F_{OLD}^{CMA} = 0.646$.

B. Modus Operandi

The following algorithmic variants have been applied to the problem:

- NSGA-II: The classical variant by Deb [12], [18].
- Metamodel-Assisted EA with Probability of Improvement (PoI-EMOA).
- Metamodel-Assisted EA with Expected Improvement (ExI-EMOA).

The parameterizations of these methods is $\mu = 50$, $\nu = 0.2 \cdot \lambda$, with two different settings for $\lambda$: $\lambda = 250$ and $\lambda = 50$. The parameters of the mutation operator and recombination operator have been chosen as described in Deb [12]. The source code used in the experiments can be retrieved from the authors on request.

C. Numerical Results: Multi-criteria Optimization

In this paper, the advanced EMOA have been applied for the first time on molecular alignment problems, and we will now give a detailed presentation of results. Note, that tests of the metamodel-assisted EA on artificial test problems have been performed in [1].

Fig. 6 to Fig. 8 display results of the test runs. The 20%, 50% (median), and 80% attainment surfaces are given. Each
one of them refers to 5 runs with 20000 evaluations per run. In order to make the curves easier to be distinguished, we zoomed in a box around the knee point of the Pareto front approximations (Fig. 9 - Fig. 11).

The results clearly indicate that there is a conflict between the two objectives, as suspected by the physicist’s intuition. Thus, Pareto optimization is an appropriate tool for solving this problem. The ‘bright side’ of the results is that a convex Pareto front has been observed. This suggests that good compromise solutions are likely to be found. We observe a sharp increasing flank at both ends of the approximated Pareto front. Regions of fair trade-offs range from about $-0.6$ to $-0.4$ (in the $(-f_1)$ coordinate).

There are significant differences in the behavior of the multi-criteria EA variants. The best coverage of the Pareto front has been achieved by the ExI-EMOA. This variant is the only variant that found solutions for $f_1$ above 0.58. The highest value found was 0.6184. The PoI-EMOA resulted in a less well-spread approximations. However, the precision of this EMOA was better in the regions covered. This results is consistent with the theoretical findings as reported in [1] and the tests on artificial problems reported there. The expected improvement measure emphasizes on exploring unknown regions, while the probability of improvement tends to better exploit regions. Note, that the metamodel-assistance seems to be a valuable ingredient for this problem, as can be seen by comparing the results of the NSGA-II with those of the metamodel-assisted EMOA.

A more detailed analysis of the metamodel-based approximations was performed, in order to assess whether the metamodelling worked in the way we expected it from theory. The results are displayed in Fig. 12 - Fig. 15 for one of the runs with the ExI-EMOA ($\lambda = 250$). The $y - \hat{y}$ plots indicate that in the whole range of function values the results obtained with the metamodel were strongly correlated with the true function values. The error bandwidth for $f_1$ is about 10% of the range for $f_1$ and 15% for $f_2$. These results correspond to results in similar studies in metamodel-assisted optimization [1]. Moreover, the lower confidence bounds are named $y_{lb}$ and have been compared to the outcomes $y$ of the precise evaluations. Here, the $95.45\%$-lower confidence bounds, as computed by the Kriging method, have been checked for their validity (Fig. 14 and Fig. 15). The results are in conformity with the theory for $f_1$. For $f_2$ some outliers in the region of $y_{lb}$ from 0.15 – 0.2 need to be reported. The reason for these outliers is not clear yet and needs to be further investigated. However, these outliers seems not to significantly harm the algorithms performance.

From the physics point of view the result is interesting, since it shows the nature of the trade-off between the angular alignment and the intensity, which is expressed here through the second harmonic generation. The importance of the intensity criterion is likely to govern the decision of the expert on the trade-off surface, which is to look for solutions with relatively good $f_1$ values in the region of fair trade-offs.
Fig. 9. 20% A.S.: ZOOM-IN.

Fig. 10. Median A.S.: ZOOM-IN.

Fig. 11. 80% A.S.: ZOOM-IN.

Fig. 12. $y - \hat{y}$-plot for $(-f_1)$.

Fig. 13. $y - \hat{y}$-plot for $f_2$.

Fig. 14. $y - y_{lb}$-plot for $(-f_1)$. 
V. SUMMARY AND CONCLUSION

There are two main results of the study: firstly, further evidence has been added to the usefulness of metamodelling in multi-criteria optimization with time-consuming evaluations. In particular, the expected improvement criterion seems to have demonstrated its potential. Secondly, with this study for the first time it has been shown experimentally that the dynamic molecular alignment problem can be considered as a multi-criteria problem with conflicting objectives, which exhibits a convex Pareto front. This is a strong result for the research of this problem domain, indicating that multi-criteria methods should be considered in the future also for similar problems. However, none of the EMOA variants, though some runs were close to the ideal points in the future.

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