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Evolutionary Optimization of Three-Photon Absorption in Molecular Iodine

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***Abstract*— We report on the application of an evolutionary algorithm to a noisy, dynamic optimization problem in chemistry: the maximization of three-photon absorption in molecular iodine. An evolution strategy is used in real-time in a closed loop experiment to search the space of physically realizable phase-modulated femtosecond laser pulses. The probability of three-photon absorption is estimated by measuring UV fluorescence. With the evolutionary search it is possible to enhance the UV fluorescence by a factor of 3.4 compared to the most intense pulse.**

I. Introduction

It has long been a goal of photochemistry to control the quantum state of a molecule with a tailored laser pulse [1]. This is known as coherent control. This amounts to finding the time-varying electric field (laser pulse), $E(t)$, that maximizes the probability of a particular quantum transition in the molecule. The dynamics of any molecule under an electric field are described by the time-dependent Schrödinger equation. Use of the equation requires knowledge of the Hamiltonian for the molecule. However, complete specification of the Hamiltonian has only been achieved for the simplest cases, notably hydrogen and some diatomic molecules. Ref. [2] proposed instead to use an optimization algorithm in a closed loop to optimize the probability of the quantum transition. There has been much success since in the application of evolutionary algorithms to coherent control of a range of optical [3], chemical [4] and biochemical processes [5]. See [6] for a review. However, there often is little discussion of the choice of objective function and the implications thereof for optimization. An important exception is [7] in which the effect of the objective function on the robustness of the optimization is investigated. Ref. [8] gives a didactic presentation of evolutionary algorithms targeted at the quantum control community together with a case study and highlights some practical considerations.

II. Nature of the Optimization Problem

The target system under consideration is molecular iodine, I_2, in the gas phase. If I_2 is irradiated with light of central wavelength 570 nm then it may absorb three photons and be excited to a higher energy level [9]. The molecule then emits a photon of wavelength 340 nm as it decays to its ground state. The molecule can be thought of as traversing a path along coupled potential energy surfaces (PESs). Coherent control of a quantum phenomenon amounts to finding a shaped laser pulse that determines the path the molecule takes along the PESs, such that the probabilty of that phenomenon is maximized [10]. We aim to find a shaped laser pulse that maximizes the probability of three-photon absorption (3PA).

A. Parametrization of Laser Pulse

The pulse is parametrized with respect to angular frequency, $\omega = 2\pi c/\lambda$, where c is the speed of light in a vacuum and λ is wavelength. The spectral amplitude window is a super-Gaussian with a full-width at half maximum of 46 nm and a central wavelength of 570 nm. The pulse is shaped by varying the phase, $\phi(\omega)$, at a number of control points, $\omega_1, \ldots, \omega_d$. The number of control points is determined by the spectral resolution of the pulse shaper. The pulse shaper is a Dazzler from Fastlite[1], this is an acousto-optic programmable dispersive filter (AOPDF) [11]. The AOPDF uses a crystal of tellurium dioxide (TeO_2) as a programmable diffraction grating. TeO_2 is birefringent, i.e. light travels at different speeds along different directions (the fast ordinary axis and the slow extraordinary axis). Light incident along the fast axis can be diffracted to the slow axis by compression of the crystal, a property known as photoelasticity. Each frequency component of the incident light can therefore be retarded by a specific amount by varying the point of diffraction. The amount of retardation for each frequency, ω, is known as the delay, $\tau(\omega)$. This controls the phase since $\tau(\omega) = d\phi(\omega)/d\omega$. The point of diffraction of each frequency is varied by varying the compression in the crystal, i.e. by propagating an acoustic wave through the crystal.

Non-linear optical processes, such as 3PA in iodine, are unaffected by the addition of $a\omega + b$ to $\phi(\omega)$ [6]. An

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optimization of $\phi(\omega)$ directly thus results in two-dimensional infinite redundancy in the search space. For this reason, we optimize the second derivative of the phase w.r.t. angular frequency, $\delta(\omega) = d\tau(\omega)/d\omega = d^2\phi(\omega)/d\omega^2$. Since there are only a finite number of control points, $\omega_1, \ldots, \omega_d$, at which the phase can be specified, we optimize $\delta(\omega_i), i = 1, \ldots, k$ and numerically integrate twice to obtain the phase. The range of $\delta(\omega_i)$ is the same for all ω_i and is determined by the physical properties of the tellurium dioxide crystal.

There are 178 evenly spaced control points from $\omega_1 = 3.45\ \mathrm{rad\ fs^{-1}}$ to $\omega_{178} = 3.18\ \mathrm{rad\ fs^{-1}}$, corresponding to $\lambda_1 = 547\ \mathrm{nm}$ to $\lambda_{178} = 593\ \mathrm{nm}$, with $\delta(\omega) \in [-7300, 7300]\ \mathrm{fs^2\ rad^{-1}}$.

B. *Fitness Function*

The iodine molecule can absorb three photons via resonant transitions. It then decays to its first excited state and emits a photon. The emitted photons (fluorescence) can be detected in a photomultiplier tube (PMT). The PMT signal measures the number of I_2 molecules that have been excited to the desired state by the absorption of three photons. This signal is, therefore, expected to be proportional to the cube of the intensity of the laser pulse. This is independently measured by a photodiode (PD) placed after the absorption cell. (Because the sample is optically thin any absorption in the molecular beam is negligible.)

The PMT and PD signals are both corrupted by random experimental noise and dark signal. The dark signal is a noisy systematic offset with non-constant mean.

We aim to maximize the probability of 3PA for a shaped laser pulse with a given power. The most common choice of algorithm in the quantum control literature is an evolutionary algorithm [12] as originally suggested by [2], although other search algorithms have been used, such as the modified simplex algorithm in [7]. These are all optimization algorithms that aim to maximize (or minimize) some objective function. In the field of evolutionary optimization the objective function is called the fitness function in analogy with Darwin's concept of biological fitness. The choice of fitness function will affect the properties of the solution found, although it is rarely discussed in the quantum control literature.

The most obvious choice of fitness function is $f_1(\delta) = \mathrm{PMT}(\delta)$, where $\mathrm{PMT}(\delta)$ is the fluorescence signal from the photomultiplier tube and δ is shorthand for the laser pulse with spectral phase obtained by twice numerically integrating $\delta(\omega_i), i = 1, \ldots, k$. However, the mean laser power is not constant during the course of an optimization owing to changes in ambient temperature and humidity that are difficult to fully control. The result is that $f_1(\delta)$ has non-constant mean for a fixed δ.

In principle, $\mathrm{PMT}(\delta) \propto \mathrm{PD}(\delta)^3$, which suggests $f_2(\delta) = \mathrm{PMT}(\delta)/\mathrm{PD}(\delta)^3$ as the fitness function. Owing to scattered light and dark signal in the PMT and PD, the proportionality does not hold in practice, so that $f_2(\delta)$ also has a non-constant mean for a fixed δ. Thus the problem of a dynamic fitness function is not overcome when normalizing the fluorescence signal by the cube of the laser power. This can be contrasted with [5], in which coherent control of a protein in the liquid phase was investigated. In that work the number of photons absorbed was used to weight the fitness, resulting in a stable fitness function. This is not possible with our experimental conditions.

The aim is to maximize the PMT signal of a shaped pulse with a given laser power. Typically, the PMT signal for a shaped pulse is compared to that of the unshaped pulse. The unshaped pulse has $\delta(\lambda) = 0$, hence $\phi(\lambda) = 0$, for all wavelengths λ. This is known as the transform-limited (TL) pulse, δ_{TL}, and is the shortest and most intense pulse for a given power and bandwidth. An enhancement of 3PA above that achievable with the TL is indicative of optical effects that are not due to intensity variation and suggests that coherent control of the optical process has been achieved [9]. This suggests $f_3(\delta) = \mathrm{PMT}(\delta)/\mathrm{PMT}(\delta_{\mathrm{TL}})$ as the fitness function. Since the laser power varies, $\mathrm{PMT}(\delta_{\mathrm{TL}})$ should be measured frequently enough that the long-term drift in the laser power is negligible. Once per generation is sufficient for the algorithm used here in the current experimental conditions. (If $\mathrm{PMT}(\delta_{\mathrm{TL}})$ were measured for every shaped pulse, then the number of fitness evaluations possible in the given available time would be halved.)

Since the mean of the dark signal varies, we also record the dark signal, $\mathrm{PMT}(0)$, prior to evaluating $\mathrm{PMT}(\delta_{\mathrm{TL}})$. The enhancement of 3PA over that of the TL for a shaped pulse, δ, is then estimated as $(\mathrm{PMT}(\delta) - \mathrm{PMT}(0))/(\mathrm{PMT}(\delta_{\mathrm{TL}}) - \mathrm{PMT}(0))$. We do not use this as the fitness function since the dark signal is independent of the laser power and the pulse shape, whereas the noise in the dark signal would increase the noise in the fitness function and lead to a deterioration in performance.

C. *Experimental Conditions*

A schematic of the experimental set-up is given in Fig. 1. Nominally transform-limited pulses of length 30 fs, centred at 570 nm from a commercial non-collinear optical parametric amplifier (Clark-MXR, NOPA) pumped by a 1 kHz repetition chirped pulse amplifier (Clark-MXR, CPA-2010) are directed into an AOPDF of length 25 mm. The shaped pulse is loosely focused with a parabolic mirror into a cell containing about 100 N $\mathrm{m^{-2}}$ of I_2 vapour at around 310 K. The resulting UV fluorescence is detected through a discriminating spectral filter at right angles to the laser propagation direction by a PMT. In order to avoid saturation effects the incident pulse energy was restricted to be less than 170 nJ. Supporting evidence that these conditions are in the perturbative regime was obtained by measuring the intensity dependence of the UV fluorescence signal, which was confirmed to be approximately cubic.

There is one further consideration when evaluating optimized pulse shapes. The AOPDF achieves phase-modulation by longitudinal dispersion of the frequency spectrum. That is, different frequencies are diffracted at different distances into the crystal and thus undergo more or less dispersion. The result is that different pulse shapes have different diffraction efficiencies, dependent on how far the various frequency

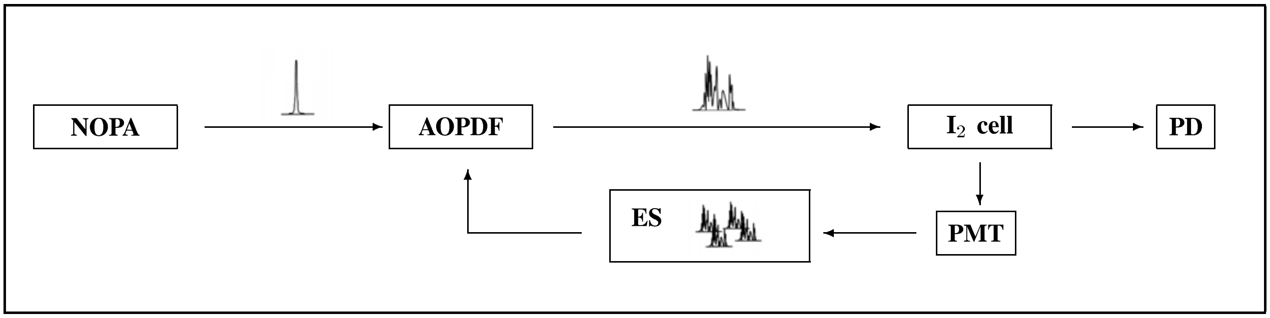

Fig. 1. Closed-loop optimization of pulse shape. An unshaped laser pulse from the NOPA is directed into the pulse-shaper (AOPDF). A pulse shape is specified by the evolution strategy (ES). The shaped pulse is focused into a cell containing iodine. The fluorescence is measured by a photomultiplier tube (PMT). The PMT signal is used as feedback for the optimization (fitness). The intensity of the diffracted pulse is monitored with a photodiode (PD).

components have propagated through the crystal. Two pulses with the same incident laser power having different diffraction efficiencies will not have the same power at the point-of-experiment. That is, the number of photons available for absorption will vary. This is partially corrected for by calibration of the AOPDF [13]. Some variation in diffraction efficiency is still evident, though. We measure the diffracted power of the shaped pulses to assess whether the variation in PMT is due to coherence effects or power variation. The diffracted power is measured by the PD simultaneously with the PMT signal.

D. *Evolutionary Optimization*

Evolutionary optimization is inspired by biological evolution [14]. A collection of potential solutions is operated on by evolutionary operators with the aim of finding an optimal solution. In analogy with biological evolution, a potential solution is called an individual and the collection of individuals is called the population. In biological evolution, the fittest individuals in a given environment are those that survive to reproduce and pass on their genetic information. This is reversed in evolutionary optimization. A fitness function is specified and the individuals that survive are those with the highest fitness.

There is an increasing variety of evolutionary algorithms (EAs). Here we consider the evolution strategy (ES). In particular, we use the standardized $(\mu/\rho \overset{+}{,} \lambda)$-ES as described in [15]. The parameters are the number of parents, μ, the number of children, λ, the number of parents per child, ρ, and the selection strategy ('plus' or 'comma'). A variety of EAs have been employed in the quantum control literature, rarely with any justification of the particular variant selected, and often with insufficient information to reproduce the experiment. We aim to introduce the optimization problems in quantum control to the EA and wider computational intelligence communities whilst arguing for a more systematic application of EAs in quantum control. In the following we detail and motivate our algorithmic design choices. There is plenty of scope for further research in identifying the optimal choice of settings for the $(\mu/\rho \overset{+}{,} \lambda)$-ES as applied to quantum control.

Briefly, the $(\mu/\rho, \lambda)$-ES proceeds as follows. A collection of μ individuals, termed parents, is selected at random from the space of candidate solutions. The fitness of these individuals is evaluated. A new collection of λ individuals, termed children, is generated by repeated application of the evolutionary operators of recombination and mutation. The fitness of these λ individuals is evaluated, and the μ individuals with the highest fitness are selected. This constitutes one generation, the best μ individuals become the parents of the next generation. The process is then repeated with the new parents until some stopping criterion is met.

The $(\mu/\rho + \lambda)$-ES is the same except for one important difference. The parents of the next generation are selected from both the λ children and the μ parents of the current generation. This is termed elitist since it is possible for a very fit individual to remain in the population indefinitely. The (μ, λ) strategy is preferred for unbounded search spaces [16], whereas the $(\mu + \lambda)$ strategy is preferred for discrete, finite search spaces [17]. Theoretically, the space of phase-modulated pulses is continuous and bounded. In practice, it is discrete and finite. It is not clear which strategy is optimal for quantum control. We prefer elitism since the algorithm is then likely to converge to a local or global maximum and we are interested in investigating the nature of the search space, and the number and nature of its maxima [12].

The $(\mu + \lambda)$-ES with a noisy fitness function suffers from over-valuation [18]. That is, the parents will have optimistically high fitness scores since they were selected as maximizing a noisy function. To avoid keeping over-valued parents, the fitness of the parents is reevaluated along with that of the children. This is particularly important in the case where the mean fitness of a given individual is drifting from generation to generation (which is the case here).

With a noise-free fitness function, elitism guarantees a monotonic increase in the fitness of the best individual from generation to generation. However, this is not guaranteed for a noisy, dynamic fitness function.

The PMT signal is noisy, which results in a noisy fitness function. Noise in the fitness function reduces the quality of the solution at convergence [19], [20]. To increase the signal-to-noise ratio, the signal is averaged over $(N =)500$ laser shots (i.e. 500 ms). The relative noise of the averaged signal is *ca.* 5%. There are more efficient strategies for sampling in the presence of noise, e.g. [19]. These are not investigated here.

Ideally, the algorithm would be run for as many generations as were required for convergence. In practice, the number of fitness evaluations is limited by the amount of time for which the laser can be operated continuously. There is a trade-off between the number of generations, n, the population size, $\mu+\lambda$, and the sample size, N [21]. We have found empirically that an (8+32)-ES outperforms a (4+16)-ES allowed the same number of fitness evaluations (and so double the number of generations). The latter outperforms the $(1+1)$-ES, which is a point-based search equivalent to stochastic hill-climbing [14]. We use the $(8+32)$-ES in the following with $n = 50$, although there is much room for investigation of the trade-off between n, μ, λ and N.

The other evolutionary operators (i.e. apart from selection) are recombination and mutation. These are designed to mimic the eponymous biological processes, see [14], [15] for details. The mutation operator is the primary search operator in an ES. The parameters controlling the mutation strategy are themselves evolved, one of the main advantages of the ES. We use intermediate recombination on the objective parameters, i.e. a child is formed by taking the arithmetic mean of two individuals randomly chosen (with replacement) from the collection of parents (so that $\rho = 2$). We use panmictic intermediate recombination on the strategy parameters [16]. We use anisotropic but uncorrelated mutations. The hyperparameters for the evolution of the strategy parameters are set using the heuristics quoted in [14].

It should be clear from the preceeding that there is plenty of scope for investigation of mutation and recombination, as well as the design of evolutionary operators for specific optimization tasks.

E. Comparison with Previous Work

Enhancement of 3PA in molecular iodine was first demonstrated in [9]. The phase, $\phi(\omega)$, can be written as a Taylor expansion in the frequency domain, $\phi(\omega) = \phi(0) + \phi'(0)\omega + \phi''(0)\omega^2 + \ldots$, where the central frequency of the pulse has been set to zero. The term $\phi''(0)$ is the linear chirp. The transform-limited (TL) pulse has $\phi'' = 0$. Ref. [9] demonstrated in an open-loop experiment that the 3PA yield has a non-linear dependence on the linear chirp. At the wavelength of 570nm, they observed a threefold enhancement in the 3PA yield for a positively chirped pulse relative to the TL pulse.

Varying the linear chirp $\phi''(0)$ is equivalent to setting all $\delta(\omega_i) = k$ in the current parametrization and varying k. This is single-parameter control. Preliminary work indicates that 3PA can be further enhanced by also varying the quadratic chirp [22]. In contrast, we aim to achieve a higher enhancement by allowing much more flexibility in the pulse shape. The downside is that it is not possible to evaluate all shaped pulses. We therefore use an evolutionary algorithm to search the space of shaped pulses.

In practice, the enhancement achievable varies with the experimental conditions. In order to provide a direct comparison with [9], we repeat that work as follows. Each generation of the ES has 40 pulses, for each generation we evaluate 40 pulses with linear chirp equally spaced in {-2281.5, 2281.5} fs^2 rad^{-1}. The fitness evaluations are interleaved with randomized design so that the experimental conditions are the same for the two experiments. We find that the optimum linear chirp is near 400 fs^2 rad^{-1} and gives an enhancement by a factor 3.0 ± 0.3 over the TL pulse. There is also a local maximum near -640 fs^2 rad^{-1}. These results are in close agreement with [9].

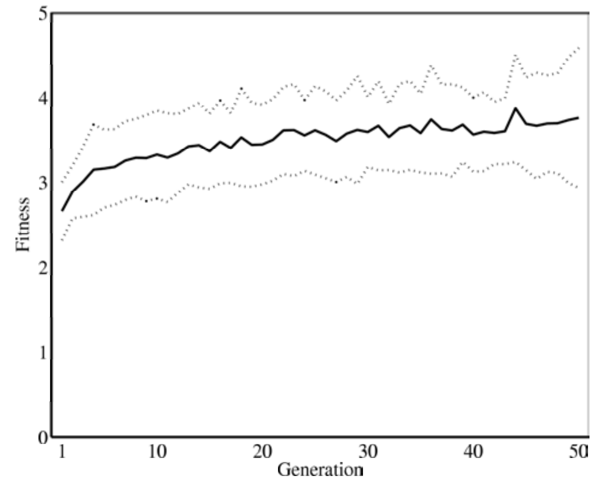

Fig. 2. Performance of the ES. The fitness of the best μ pulses in each generation is shown. The fitness is averaged over 16 successive runs. The dotted lines indicate one standard deviation.

III. Results

The fitness of the ES through the evolutionary run is shown in Fig. 2. We plot the mean fitness of the best $\mu(= 8)$ pulses each generation. This is averaged over 16 successive runs of the ES (four per day on consecutive weekdays). Results are reported as the mean $\pm$ one standard deviation over the 16 runs. The initial fitness is 2.7 ± 0.4, this corresponds to an enhancement of 3PA by a factor of 2.6 ± 0.3. The initial fitness is the mean fitness of the fittest eight pulses in the initial random sample of 40. That this is so high is due to a peculiarity of the parametrization. By the central limit theorem, a random sample of pulses from $\delta \in [-7300, 7300]^{178}$ has an average linear chirp (over ω) with distribution $N(0, 316^2)$. Since there are local maxima w.r.t. linear chirp at $\phi'' = 400$ and $\phi'' = -640$, some of the randomly chosen pulses will already have near optimal enhancement.

The ES gradually improves on the initial fitness throughout the optimization. The average fitness in the final generation is 3.8 ± 0.9, corresponding to an enhancement of 3PA by a factor of 3.4 ± 0.4. There is wide variation in the performance from day to day, owing to uncontrollable changes in experimental conditions. The greatest average enhancement on convergence was by a factor of four. On all but two of the 16 runs, the average enhancement of 3PA of the best μ pulses in the final generation was higher than that of the best linearly chirped pulse.

An examination of the fitness on the separate runs suggests that in some cases the ES had not converged. Preliminary results with $n = 100$ generations, however, do not improve on the results obtained with $n = 50$ generations.

The diffracted power of the optimized shaped pulses was the same as that of the transform-limited pulse. This indicates that the enhancement in 3PA is not due to intensity effects.

IV. Conclusions

We have reproduced the result of [9], *viz.* that three-photon absorption in molecular iodine can be enhanced by a factor of three compared to the unshaped transform-limited pulse by the addition of linear chirp. Moreover, we have shown that it is possible to improve on this by using an evolution strategy to search the space of phase-modulated pulses.

Ref. [9] identified two local maxima of 3PA when varying linear chirp. Ref. [22] showed that these can be improved by also varying the quadratic chirp and identified a third local maximum with that parametrization. Since the initial population is likely to contain pulses close these maxima, it is possible that the ES is fine-tuning the previously found pulse shapes. Characterization of the optimized pulse shapes should identify if this is the case, or if the ES has found entirely novel pulse shapes.

V. Future Work

As discussed in the text, there are a number of design choices that have not been fully investigated. These include the population size, the selection strategy, the number of generations and the sample size, as well as the fitness function. For example, initial results suggest that increasing the number of generations is not beneficial.

We have argued that the appropriate fitness function is $f_3(\delta) = \mathrm{PMT}(\delta)/\mathrm{PMT}(\delta_{\mathrm{TL}})$ since $f_1(\delta) = \mathrm{PMT}(\delta)$ is dynamic. However, preliminary results with the same ES maximizing $f_1(\delta)$ suggest that the latter is preferable. Fitness function $f_3(\delta)$ is estimated from two noisy signals whereas $f_1(\delta)$ contains only one noisy signal. It seems that the advantage gained in having a fitness function with constant mean is outweighed by the increased noise. Further investigation and signal analysis will shed light on this.

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