Robust Basis Functions for Control from Dimension Reduction of Adaptive Pulse-Shaping Experiments

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Abstract. A new statistical analysis of our adaptive control experiment reveals that only seven simple basis functions are needed to account for ~ 90% of the fitness variance in 11700 laser pulses explored during a 208-parameter adaptive optimization.

1. Introduction

The adaptive femtosecond pulse shaping methodology first proposed by Rabitz[1] offers a powerful tool for selectively manipulating complex molecular systems with light. This has lead to the discovery of control fields that are difficult to predict a priori[2-6]. However, understanding how to infer control mechanisms from these fields remains an outstanding question that must be addressed if the technique is to become a general tool for investigating chemical dynamics.

In many adaptive pulse-shaping experiments, astronomically-large search spaces are made possible by manipulating large numbers (on the order of 100) of frequency-dependent parameters such as phase and amplitude. In some reports, adaptively-discovered laser fields reveal regular structure such as evenly spaced pulse-like features[4, 5]. In these cases, mechanistic insight has been gained using parameterization techniques that recreate salient pulse characteristics combined with pump-probe experiments that test the adaptively discovered features. However, the massive parameter space that facilitates control in the first place can also serve to obscure mechanistic insight. This is because the heuristic optimization algorithms do not distinguish between pulse features necessary for control and those that contribute negligibly to fitness[7, 8]. In order to understand control mechanisms it is important to distill adaptively-shaped pulses to their essential control structure using an intellectually tractable number of variables. Consequently, much recent research has been directed toward dimension reduction of adaptive control results[8-11]. Our analysis suggests that statistical methods can pay too much attention to the mechanism of the search algorithm rather than mechanisms of control. To overcome this, it is important to quantify the correlation of phase-parameter variance with experimentally measured pulse fitness.

2. Results and Discussion

The experimental setup we use is similar to one previously described by Gerber's group[12]. In our experiment, the algorithm was run for 195 generations (11700 total pulses) and the optimal pulse was found to approximately double the emission/SHG ratio with respect to randomly encoded pulse shapes.

The adaptive algorithm manipulates the phase modulo 2π and the resultant phase functions for all but the simplest laser fields can exhibit discontinuities between adjacent pixels. For statistical modeling purposes this is problematic because the relative phase, rather than the absolute phase, determines pulse fitness. After implementing an 'unwrapping' algorithm, the normalized covariance matrix can be calculated and diagonalized to reveal the fundamental directions along which phase functions vary in the total data set. These are known as principal components (PC's). White *et al*. have argued that the few PC's of statistical importance (highest percentage of the variance) reveal the fundamental degrees of freedom in the control Hamiltonian[11].

While variance does contain information about the fitness, their relationship is necessarily obscured by the heuristic nature of the evolutionary algorithm's search mechanics. Consequently, we have developed a general analysis tool[13] that is based on partial least squares regression (PLS) of the normalized covariance matrix[14]. The regression algorithm fits the phase variables to a minimum dimensional hyperplane in the search space. The plane is expressed with an orthogonal basis set such that the first basis vector points along the direction of greatest fitness variance on the hyperplane, the second basis vector points along the direction of next greatest variance, and so forth. This reveals the fundamental directions wherein changes to the phase functions correlate most strongly to fitness. These orthogonal vectors are linear combinations of the original 208 phase variables. The extracted vectors are quantified in terms of both the percentage variance in phase as well as fitness. Seven orthogonal inter-pixel phase relationships account for 89% of the observed fitness variance (Table 1). It is interesting to note that the third PC accounts for 11% of the fitness variance despite exhibiting negligible phase variance. This would have been overlooked had we only considered the diagonalization of the covariance matrix.

Table 1. Phase and fitness variance for PLS dimensions.

PLS Dimensions	*% variance in phase*	*% variance in fitness*
PC 1	62	47
PC 2	31	24
PC 3	1	11
PC 4 - PC 7	≤ 2	≤ 2

Each of these basis vectors can be plotted in terms of relative phase versus pixel number. If the phase unwrapping procedure is neglected prior to the PLS analysis, highly discontinuous functions are discovered. With phase unwrapping prior to PLS analysis the extracted basis functions are remarkably simple as shown in Fig. 1(a). Optimizations that utilize few simple variables discovered during PLS analysis of larger data sets are highly efficient as shown in Fig. 1(b).

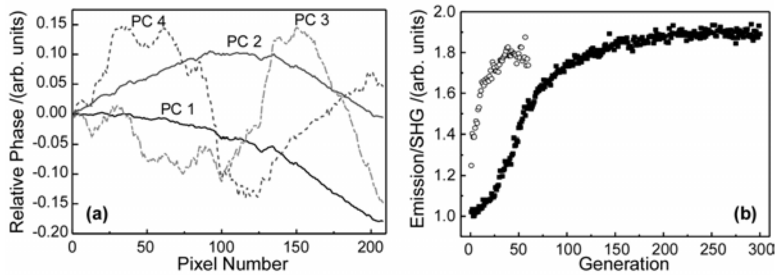

Fig. 1. (a) PC's 1 – 4 revealing the interpixel phase relationships accounting for 84 % of the fitness variance. **(b)** (¦) An optimization curve for a 210-variable ratio experiment. (?) A curve where only *eight variables* discovered from PLS analysis of the larger set were used.

The small number of simple functions needed to account for the majority of the fitness reveals that highly structured pulse features are not necessary for increasing emission/SHG in this experimental system. This is consistent with a control mechanism that exploits broad features in the two-photon absorption spectrum of the molecule as suggested by the Gerber group[12] and the Joffre group[15]. Most importantly, the analysis reveals *in a completely general fashion* which and how many independent experimental "knobs" are needed to manipulate control mechanisms unveiled by the larger adaptive search.

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