Implementing Optimal Control Pulses in ESR using an Arbitrary Waveform Generator

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Introduction

Microwave pulses with variable amplitude and phase can be designed, using optimal control theory, to achieve higher excitation bandwidth, and hence higher sensitivity, than hard pulses. The main problems in directly applying optimal control techniques [1] (designed originally for NMR) to EPR spectroscopy are the timescales and the spectral widths involved.

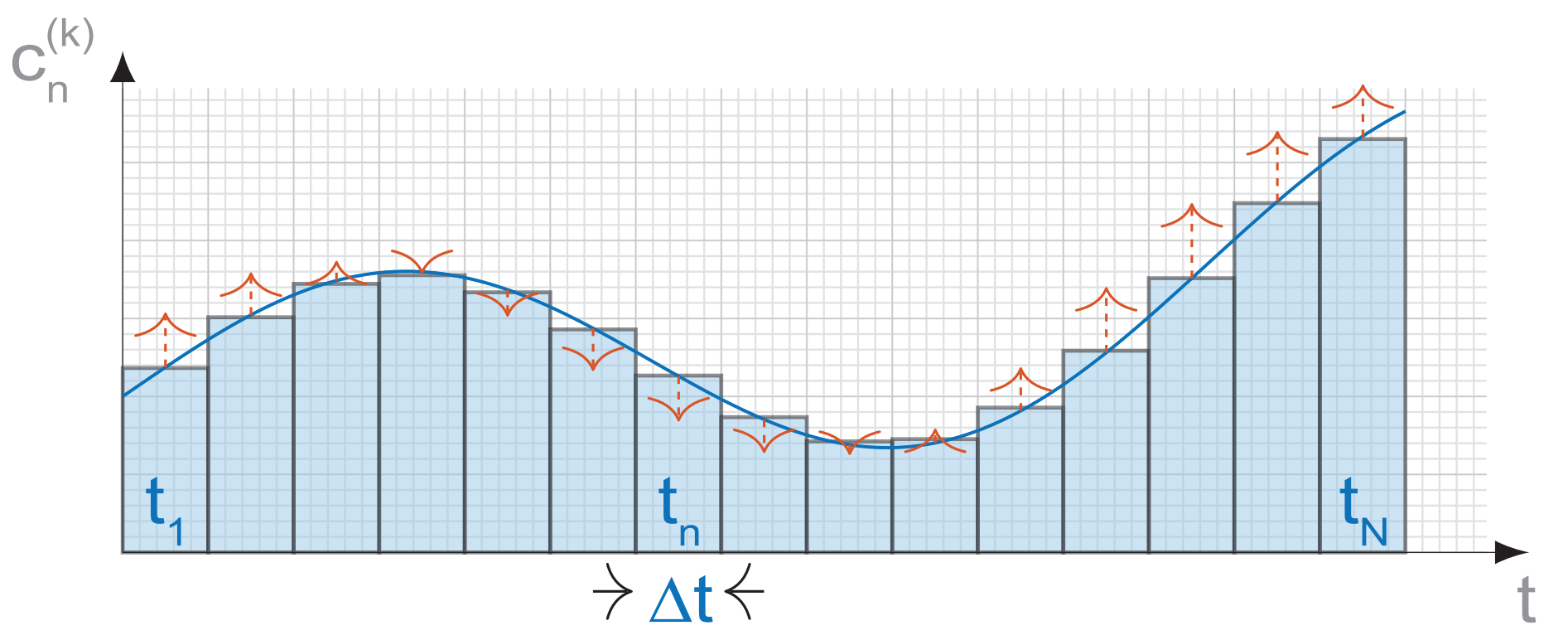
Bruker SpinJet, an arbitrary waveform generator (AWG), makes it possible to use a discretised pulse shape to realise those optimal control solutions. However, waveforms produced by an AWG inevitably become distorted in transit between the AWG and the sample. A number of solutions have been proposed for creating a transfer matrix to transform the proposed input waveform to that seen by the sample [2, 3, 4].

In this communication we propose using the ideas of previous work, using an AWG to send shaped pulses to an ESR experiment [2-8], in forming a transfer matrix from measurements at the transmission monitor, after the AWG. This transfer matrix is then used within a cost function for quasi-Newton optimisation (tolerant to static noise), then simulated with gradient ascent pulse engineering (GRAPE) [1]. Typical optimal control pulses involve sharp transients and it is expected that those would generate pronounced waveform distortions. To reduce this effect, we propose the use of smoothing Tikhonov regularisation functions within the optimisation.

The numerical solutions produced by the cost functional described above would not be optimal in the strict sense; further waveform distortions are expected to be sample- and resonatorspecific. The proposition here is to use the GRAPE solution as a starting point, close enough to a local minimiser, from which a modified simplex method, running in a direct measurement feedback loop on an actual instrument, would not struggle to find a minimiser propering form:the time steps. A particular strength of the GRAPE method is that the gradient of the fidelity functional

The piecewise-constant Hamiltonian approximation of the GRAPE algorithm [1] is essential to simulating optimal control solutions.

It is important to realise the rectangular, shaped pulses on the experimental hardware - typically, the AWG will interpret shapes as point-to-point triangular pulses, then interpolated to the characteristic bandwidth of the hardware.

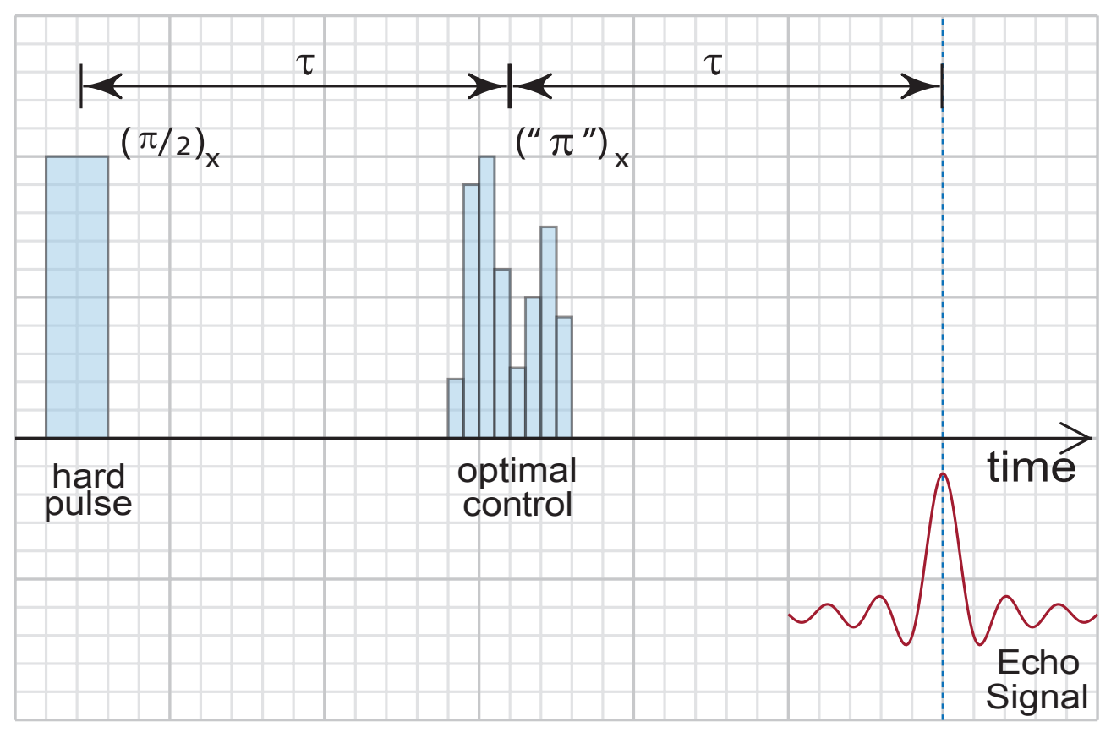


Experimental Design

As an initial "proof of principle", we test a simple two pulse echo experiment of a Finland Trityl sample. The two-pulse experiment, with a 90° hard pulse followed by a 180° hard pulse, is used throughout the optimisation as a metric of comparision.

An initial test of the optimal control is to replace one of the hard pulses by a set of GRAPE pulses. The *Spinach* sofware simulation [9] is set to simulate the Finland Trityl using an electron g-factor of 2.00270 with relaxation damping rate of 50μs. The point-to-point state transfer GRAPE simulation uses 256 time steps of 0.625ns and transfers from an initial state of $\mathbf{L}_z$ to a desired state of $\mathbf{L}_+$. The simulation states from an initial random guess.

The optimisation algorithm is a quasi-Newton-BFGS - and the optimisation takes a very small number of iterates (considering the simplicity of the system).

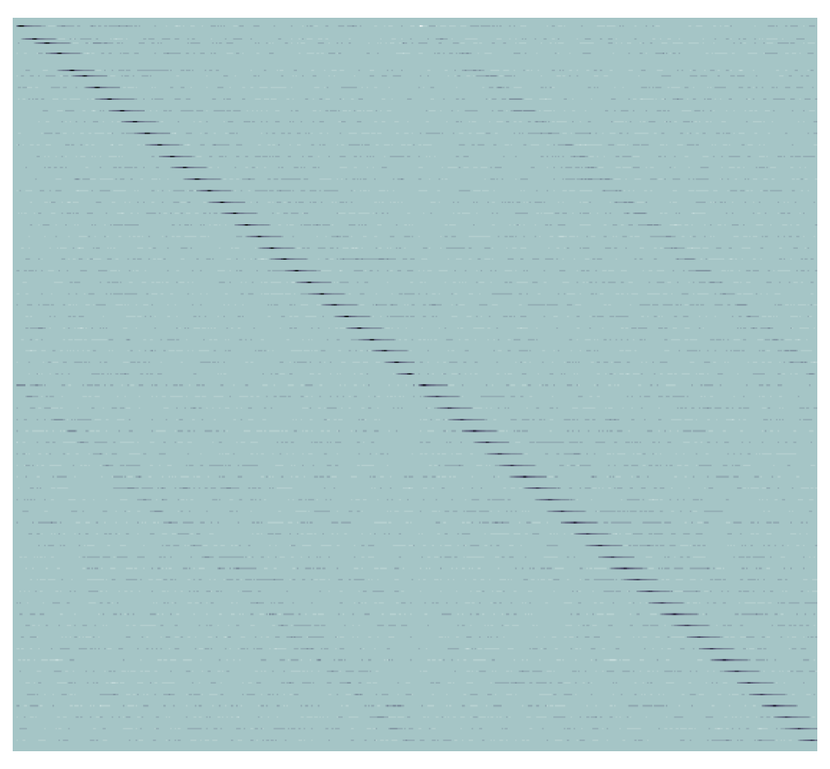


The Transfer Function

Any electrical system will suffer a level of "noise" induced by environment specific interactions. This is true of ESR spectrometers - but a more fundamental contribution is a bandwidth mis-match between different parts of the hardware that make up the system,

The instrumentation in this work has a 1GHz spectrometer tranmission monitor output, and a 1.6GHz AWG unit. The AWG unit is set up to interpolate the shaped pulse it sends to the natural sampling rate of the main spectrometer hardware. This lead to a pulse distortion - particularly when using the smallest time units.

This work attempts to resolve this main issue by creating a transfer matrix, constructed from a large number of random pulse-sets, from the input to the AWG and the output from the transmission monitor.



Representation of a transfer matrix for a 32x5ns pulses (non-linear colour scale to highlight off-diagonal elements)

Feedback Control with Gradient-free Optimisation

When an optimal solution has been found analytically, it is not expected to be exactly at the experimental minimum: different laboratory environment, different background signal noise, slightly different hardware calibration - these all change the expected behaviour of the hradware. One solution is to use a gradient-free optimisation strategy, using feedback control, to guide the system to its minimum proper. Expecting this minimum to be close to the analytical minimum - a gradient-free optimisation should not be troubled. This process of using feedback control after an anylitical solution has been used successfully in other areas of physics using an AWG, and is named the Ad-HOC method [10].

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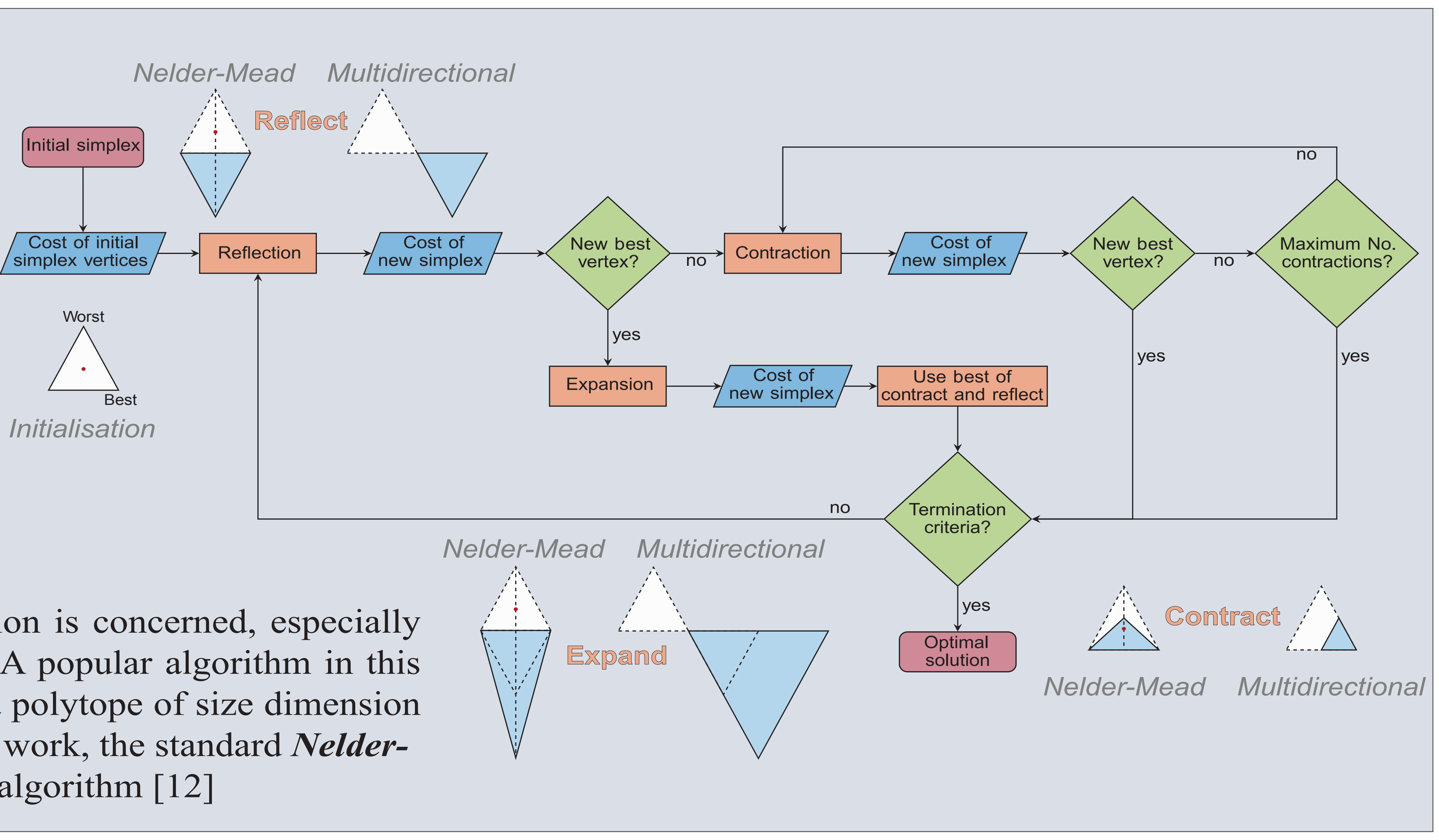
A Hybrid Simplex Method

"*There is no such thing as a free lunch*" where gradient-free optimisation is concerned, especially when a functional evaluation is expensive (performing an experiment). A popular algorithm in this context is the ***Nelder-Mead*** simplex algorithm [11], where a simplex is a polytope of size dimension n+1, n being the number of variables the optimisation can control. In this work, the standard ***Nelder-Mead*** algorithm and a parallel version of it, the ***multidirectional-searc***h algorithm [12]



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(Below):
Coherence order amplitudes representing the state transfer from Lz to L+.

Results are for the system set out in the text, simulated with GRAPE using Spinach. (optimiser results:

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==========================================================
  Optimisation Results
      Algorithm Used        : Hessian update (BFGS)
      Iterations            : 9
      Function Count        : 14
      Gradient Count        : 14
      Minimum found         : 0.35685
      norm(gradient)        : 1.2044e-09
      Total Time            : 25.3895 seconds
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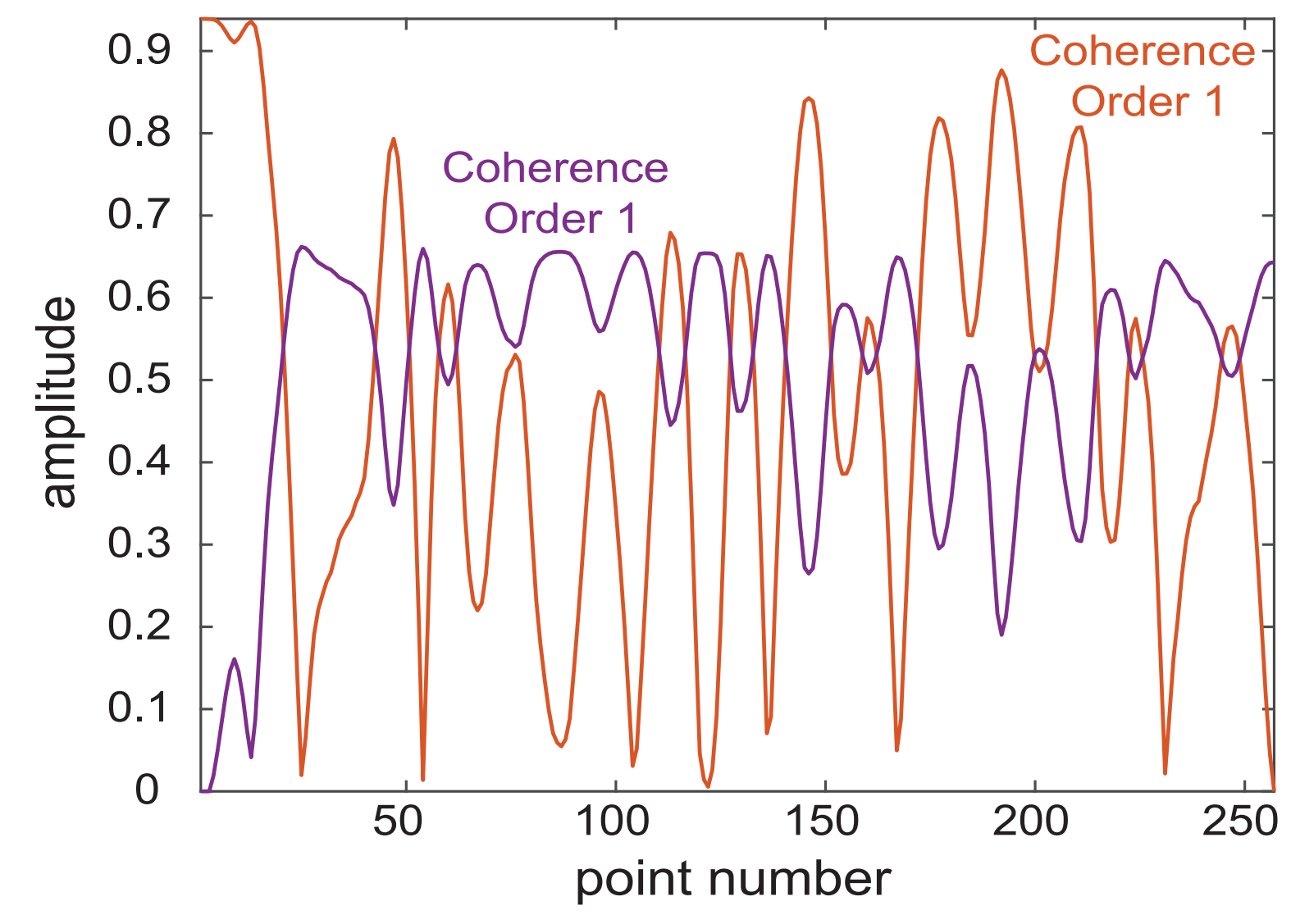

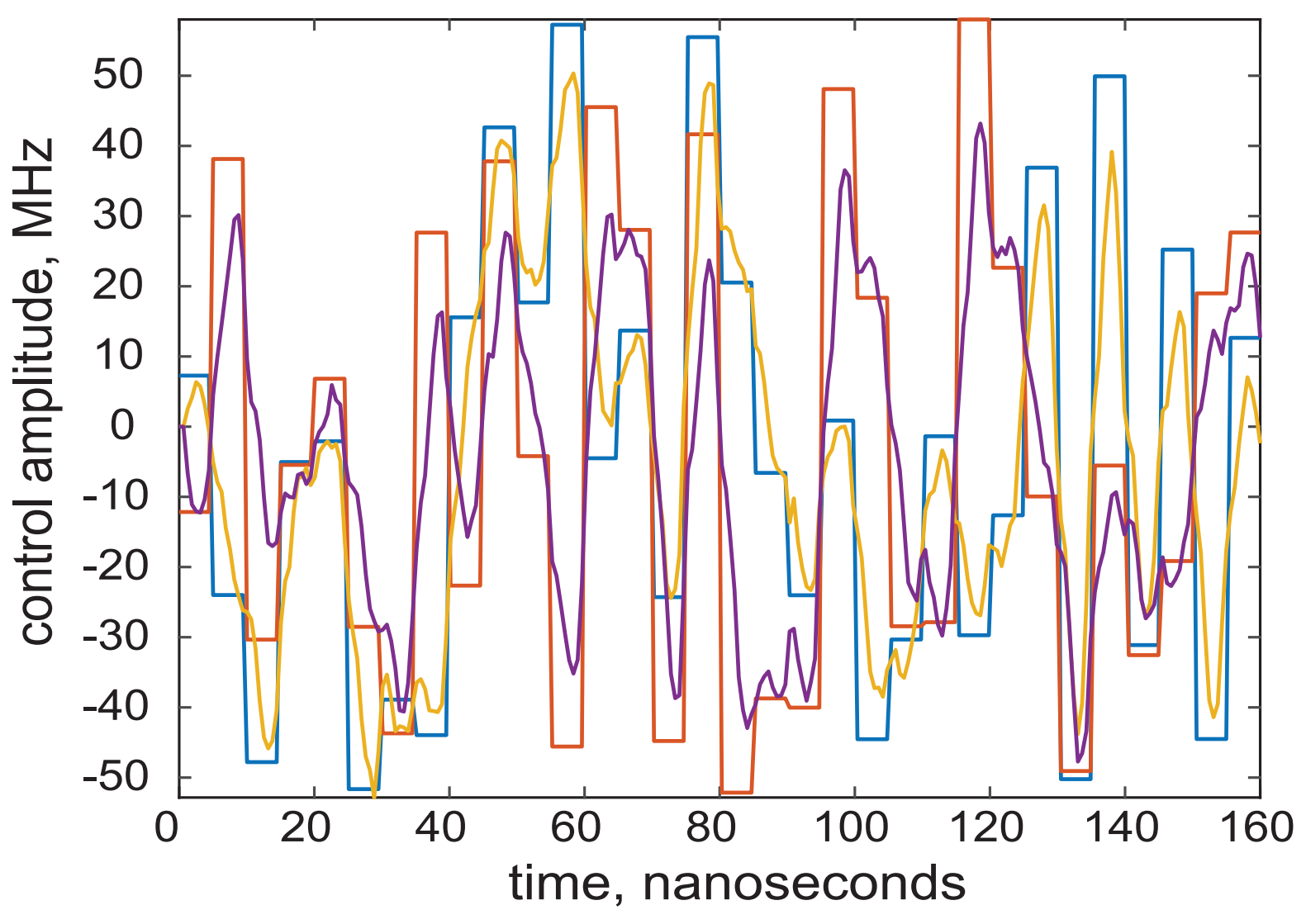

(Above):
Optimal control pulses produced by GRAPE. The plot shows the comparison between the output from the optimiser to the modified pulse set sent into GRAPE - modified with the transfer matrix shown previously (rectangular pulses each of 5ns).

(Below):
Plot showing the resulting experimenal echo using the optimal pulse set (left), for the 2-pulse echo experiment, with the second pulse replaced by the optimal set, set out above.

The solid lines represent the signal from the optimal experiment, and the dotted lines represent the signal from the standard hard-pulse experiment.

Results clearly show an improved echo integral, and a small improvement in intensity. It is expected that further optimisation with feedback control, outlined above, will further imporve results.