Dear Editor,

Given the referee comments and editorial decision, we request that our manuscript be transferred to Physical Review A as a Letter. Below we address the specific points raised by the referees and detail how we have modified the manuscript.

Thank you for your consideration.

Sincerely,

Sivaprasad Omanakuttan (for the authors)

**Responses**

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Report of Referee A -- LG18502/Omanakuttan  
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The authors do not present a novel optimal control method, as the  
robust optimal control method has been outlined in previous work. The  
authors' submitted work does not present a new theoretical advance, as  
controllability of high-dimensional, uncoupled, spin systems has also  
previously been published (notably by the authors' affiliated groups).  
I fail to see how handling the optimal control of a 10-level system is  
an advance on handling a 16-level system.

**Response:** The main result of this work is not the dimension of the Hilbert space, but the physical platform and mechanism for control. For 16-level system in cesium, the information is encoded in hyperfine levels, which includes electron and nuclear spin. This provides for certain avenues for control (e.g., microwave Rabi flopping at the hyperfine splitting), but it comes at the cost of increased sensitivity to magnetic fields. Encoding information purely in nuclear spins, and controlling spin > ½ with magnetic and optical spins is the most important innovation of this paper. Nuclear spins are highly coherent and isolated from the environment. Importantly, this can be done with very high fidelity due to the unique properties of alkaline-earth atoms, such strontium considered here, which its narrow intercombination lines, in a technology that is now well honed due to developments in optical clocks, and their application now for quantum information processing. This is the major advance of our work.

**Referee A: *Q1)*** *There would need to be extensive appendix sections explaining  
exactly how the authors calculated their pulses; simply saying "a  
modified version of grape" does not make work reproducible. I hunted  
for supplementary/appendix sections for quite some time.*

**Response**: We have added a detailed supplementary material for the paper describes how we design the waveforms using quantum optimal control. In particular we use the well-known GRAPE algorithm, based on a piecewise-constant parameterization as used in Ref. [41]. By choosing the number of steps and the phase jump between steps, we can obtain relatively smooth waveforms within reasonable time for computer optimization. We further explore how finite bandwidth of a physical controller may limit the fidelity and show how we can still obtain high fidelity in a proof of principle simple model of a low-bass filter.

**Referee A: *Q2)*** *The authors need to cite optimal control work in the wider areas of  
physics: the list of references is quite narrow and many citations are  
not the first instance of the work cited. For example, the areas of  
magnetic resonance and quantum control published much of the  
theoretical framework in optimal control in the late 1990s to  
mid-2000s*.

**Response**: We have expanded the reference list as suggested.

Report of Referee B -- LG18502/Omanakuttan  
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It is shown in the paper that a combination of the AC-Stark shift and  
optimal phase shaping can be utilized for high fidelity realizations  
of quantum states and unitary gates of a single nuclear spin in 87Sr.  
The idea is interesting, but it was proposed in previous works and  
partially demonstrated in experiments on cesium.

**Response**: We agree, but the major advance here is the application to alkaline earth atoms, which have unique properties due to their narrow intercombination lines. In addition, the technology for achieving this is now well honed due to developments in optical clocks, and their application now for quantum information processing. We believe this application is timely, and of importance to the broad community.

1. **Referee B: *Q1)*** *One of the main results of the paper is the high fidelity obtained  
within the limit set by decoherence, but this is not surprising. As  
the authors mentioned, the Hamiltonian of Eq. 1 shows that the system  
is controllable, and it follows that any state or unitary gate can be  
realized by optimal control. The order of magnitude of the infidelity  
can be roughly estimated by T/T2, where T is the duration of the pulse  
and T2 the decoherence time. I think that of interest to the  
experimentalists is not only the numerical figure of the fidelity, but  
also whether the optimal pulse can be realistically implemented with  
current rf generators. Pulses that vary too rapidly are not possible  
due to the limited bandwidth of the generator. There are also  
constraints on the maximum amplitude and minimum time step. Since the  
authors did not show any optimal pulse shape, it is hard to judge  
whether their fidelity results can be realized in experiments.*

**Response**: A key point of our result is the T2/T cannot always be large, in principle, no matter what pulses we use. In our problem the essential nonlinearity that leads to controllability is the tensor light shift (this gives the quadratic term in the Hamiltonian). However, the light induces decoherence due to photon scattering. The strength of the tensor light shift cannot be made arbitrarily large compared to the photon scattering rate. This depends on the ratio of the excited state hyperfine splitting to the natural linewidth. Indeed, in prior experiments in cesium, Ref. [43], this ratio was small, which severely limited the fidelity. A critical point is that we have a much more favorable figure of merit for the alkaline earth using the intercombination line. We think this is particularly important because arrays of optically-trapped alkaline earth atoms are prime candidates for quantum information processing and we have detailed a protocol where we can achieve very high fidelities on nuclear spin qudits, previously not seen.

Our paper details the important point above, but for a letter, this is not a detailed experimental proposal. We do include supplemental material as suggested and look at the proof-of-principle requirements to achieve pulses that are experimentally feasible. We find for modest targets, we can achieve the high fidelities near the ideal optimization with an rf-Rabi frequency of 100 Hz, and a bandwidth of 1 kHz.

**Referee B: *Q2)*** *All parameters in Fig. 2 are scaled by Omega\_rf, but the value of  
the ratio between the decoherence rate, Gamma, and Omega\_rf is not  
given.*

**Response:** Here \Gamma/2pi = 7.5 kHz and is specified in the text. The other parameters are set by the laser intensity and detuning. These determine \beta and the photon scattering rate \gamma\_s. The ratio of \beta/\gamma\_s = \kappa, which is shown in the inset to Fig. 1. Once we choose the optimal detuning, \gamma\_s is set by \beta, and \beta is set by the intensity to be optimal relative to \Omega\_{rf}

**Referee B *Q3)*** *For the robust control of unitary maps in Fig. 3 the fidelity is  
larger than 99% with a pulse duration of 10.5 ms, as claimed on page  
4. I found this result unexpected because this pulse duration is much  
longer than the decoherence time of the 5s5p excited state (2pi/Gamma  
= 0.13 ms), so it is not clear how a >99% fidelity is possible.*

**Response:** The relevant decoherence is due to optimal pumping. This occurs at the rate \gamma\_s = (s/2)\*\Gamma, where s is the saturation parameter. As we are detuned further away from resonance, we have more time than the lifetime of the excited state. In addition, working with smaller intensities (lower \beta) improves this further.

**Referee B *Q4)*** *In the numerical optimization the authors chose the average  
fidelity of 20 randomly generated states, or 10 unitary maps, as the  
figure of merit. Since the Hilbert space of a single qudit has an  
infinite number of states and unitary maps, it is not clear how  
statistically significant a set of 20 states, or 10 unitaries, is. I  
am also not sure how relevant randomly generated unitary maps are for  
quantum computation. It would be more interesting to demonstrate  
optimal control for a universal gate set, i.e., a finite set of gates  
from which any unitary can be approximated with arbitrary precision.  
An example of a universal set for single qubit gates is the Hadamard  
and the pi/8 gate (as discussed in Nielsen & Chuang). There should be  
a similar set for qudits.*

**Response:** Our goal here is to show that we could create any gates with good fidelity than focusing on a particular set of gates relevant for quantum computation. Also, a random unitary is expected to be hardest to achieve.