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PO Box 36
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# RPPR Final Report
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**INVESTIGATOR(S):**

**Name:** Herschel Rabitz
**Email:** hrabitz@princeton.edu
**Phone Number:** 6092583917
**Principal:** Y

Organization: **Princeton University**
Address: PO Box 36, Princeton, NJ 085442020
Country: USA
DUNS Number: 002484665
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Email: hrabitz@princeton.edu
Phone: (609) 258-3917
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**Major Goals:** All modern high technology machines, from aircraft out to self-driving cars, contain large numbers of (i.e., multiplexed) switches and sensors to assure high performance or alter the machine's function as desired. Similarly, an emerging high priority goal in biology is to create what may be called two-way optical communications and control with living systems, enabled by a multiplexed biological switching and sensing capability. Here, switching refers to the turning off or on of particular biological pathways or processes, and sensing refers to detection of the resultant action of the switching events. Great progress is being made using genetic engineering to create multiple classes of switches and sensor molecules, often consisting of proteins with an embedded chromophore. Typically, the chromophore undergoes an optically induced structural transformation, corresponding to either a switching event in the case of so-called optogenetics or, concomitantly, in the case of sensor molecules, the creation of a state that may be detected (e.g., by fluorescence). The recent growing family of optogenetic switches or sensor proteins addresses only half of the switching/sensing objective, as it is necessary to employ appropriate optical sources to utilize these molecules for their designed purposes. Up until the inception of the present research project, the operation of molecular switches and the observation of subsequent sensing was performed in the linear optical regime, thereby greatly limiting the number of switches and sensors which may simultaneously function in complex biological environments. Thus, to meet the clear demands for operation of high numbers of multiplexed switches and sensors, the present research introduced advanced optical broadband radiation with shaped pulse control capabilities, in order to discriminately address one optogenetic switch over another and similarly detect one protein sensor over another. Novel optical resources were developed in our basic research for this purpose with the aim of reaching breakthrough switching and sensing operations of technological significance. We have currently reached the level of simultaneous detection of three spectrally extremely similar proteins, which is already a record in the field.
The research centered around achieving high quality switching and sensing in biological molecules sits on the broad foundation of theoretical and experimental principles of quantum control. Thus, the research also had the goals of filling out particular aspects of the latter two general fundamental domains which support the biological control goals as well as have high value unto themselves in the quantum control field. In particular, this research aimed to seek fundamental principles of quantum control and the appropriate algorithms for achieving high performance outcomes. These principles ranged from considering the underlying control landscape over which optimization is performed, out to performing various experimental demonstrations of control to assure that the principles are on a sound footing. Furthermore, with an eye towards future practical considerations, the research also aimed to explore the prospect of creating flexible, broad bandwidth incoherent control resources which particularly can be effective in chemistry applications and yet cost a fraction of typical modern fs lasers. The research also aimed to explore the fact that concepts of molecular control extend beyond the use of lasers to

ultimately consider selected chemicals as controls themselves. With this in mind, a focus was given towards creating a new technology based on tailored peptides as “controls” to interdict in biological processes serving essentially as control agents serving as pharmaceuticals. Thus, we were mindful of relevant technology transfer ensuing from the research throughout its execution.

**Accomplishments:** The research during this past year addressed three topics, consisting of optogenetics, sensing, and the establishment of the means to accelerate the discovery of effective laser controls. A summary of the activities is given below.

1. Optogenetics. We explored two approaches to achieve effective, clean switching in optogenetics, consisting of either (a) a pump/dump process or (b) a Raman excitation step followed by a pump, but requiring no dump pulse. In both cases, broad bandwidth optical pulse shaping was utilized. Our initial research focused on the direct pump/dump route (a), which was performed in a sequence of iterations, such that a switch may be incrementally turned on or off by a repeated series of tailored pulses and their timing. The experiments focused on the so-called Cph8 optogenetic switch, which may be easily turned to the on state, but has defied simple optical means to be turned off. Our experiments achieved 85% switching to the “off” state, which is an unprecedented outcome. Importantly, the iterations are on a time scale much faster than any relevant biological processes, thereby forming a practical switching capability. Additionally, we successfully performed simultaneous control experiments on two laser dyes to simulate the dual vibronic transitions linked to form the overall optogenetics switching mechanism (a).

Our research also focused on adoption of the mechanism (b) referred to above, where a Raman excitation step prepares a vibrational state population in one arm of the optogenetic switch followed by a pump pulse. The net gain is potentially very significant as it has the capability to eliminate the need for optical dumping, and secondly, reduce the number of iterations involved in either the on or off optogenetic switching operations. The basic Raman principles were successfully tested in various organic molecules to demonstrate the sound nature of the concept. A final transition to biological applications of path (b) entails future research.

2. Sensing. Our earlier experiments at simultaneous detection of very similar chromophores indicated that both stability and bandwidth are critical optical resources. Thus, the goal of multiplexed molecular sensing with many extremely similar protein chromophores demanded that we assemble an entirely new apparatus to greatly advance bio-sensing capabilities. In particular, we set up an optical frequency comb as a source of very broad bandwidth stable radiation, which consists of thousands of individual phase-locked laser lines. Importantly, the comb has essentially empty space between each of its myriad of lines, which we exploited as a quiet regime for sensor detection. The empty space between the comb lines was utilized by introducing high frequency modulators to shift the detected frequency signals to lie in between the laser comb lines such that we can essentially get background-free detection. Operating in a nonlinear optical mode greatly enhanced discriminating control for detection (i.e., by manipulating controlled constructive/destructive interference to enable sensor discrimination), which was achieved by splitting the original comb beam into three separate beams, each modulated at their own incommensurate high frequency, and then the three beams were brought together in the sample for detection. Operation in this mode with a comb laser had never been attempted before, and we were pleased to see for the first time that the predicted essentially noise-free heterodyne detection could be achieved. With this advance, we were able to simultaneously discriminate between three spectrally very similar bio-chromophores. The discrimination quality was far higher than any previous efforts based on simple fluorescence detection. Furthermore, the nonlinear optical comb-based technique does not require that the chromophores actually fluoresce in order to detect a sensing signal, thus significantly reducing the chance of damage to the biological cells. Future research aims to push the limits of the sensing capability to seek the maximum number of multiplexed chromophores which may be distinguished simultaneously.

3. Fundamental Quantum Control Principles. The research aiming at biological switching and sensing rests on the use of quantum control techniques. Any quantum control experiment requires finding suitable optical pulse shapes in order to reach the highest degree of performance. These general background circumstances led to us perform a wide-ranging series of theoretical and experimental studies of value unto themselves as well as directly bolstering the prospects for creating highly functional biological switches and sensors. The first set of studies were at the basic level, considering the behavior of so-called quantum control landscapes, which correspond to the physical observable as a function of the control variables. The topology and other structural features of these landscapes directly influence the viability of practically finding effective controls in the laboratory. Key advances were made to now include more complex Hamiltonians relevant to biological systems upon consideration of control landscape behavior in classical molecular dynamics. Interestingly, quantum and classical dynamical systems have the same topological landscape features, providing a natural bridge towards large biological molecules under control, which

should have a high degree of classical character. Additionally, we were able to make a theoretical breakthrough and extend the control landscape principles to general non-linear dynamical systems. This latter work encompasses quantum and classical dynamics as special cases, but most importantly provides a critical linkage to applications throughout the sciences and beyond. A number of quantum mechanical algorithmic studies were undertaken, including the introduction of a synergistic control capability for maximally drawing on the benefits of modelling and experiments, simultaneously. Further advances were made in algorithms for accelerating many-body quantum dynamics under control. The algorithms permit assessing the scaling capabilities of the multiplexed biosensing experiments, resulting in the conclusion that there is no fundamental limit (i.e., beyond having suitable optical resources), thereby allowing us to push for large numbers of sensors and switches under control. Throughout the research, we have paid attention to technology transfer drawing on our new experimental capabilities. One example is the creation of a flexible incoherent broadband light source with pulse shaping capabilities while also being at far reduced cost compared to standard lasers used in the quantum control domain. The technology involved was filed as a patent with a very favorable initial response and the technology has been licensed exclusively to Intelligent Material Solutions. In another domain, a proprietary technology was filed with Princeton University, laying out the capability for the rapid discovery of peptides as effective “control” agents in potentially treating a host of diseases. The company Manzara Therapeutics was formed around this technology with a number of initial successes drawing the attention of the investor community.

**Training Opportunities:** The research was carried out by four postdoctoral associates (i.e., at any given time, four particular individuals were involved) and three graduate students (i.e., including a SMART scholar who finished his Ph.D. under this research), covering all aspects of the experimental and theoretical studies. The group worked as a team, such that everyone was involved in a rotating basis in the associated activities, thereby assuring the widest experience and knowledge for each of the scientists involved. We also have close interactions with molecular biologists at Princeton directly interested in our optogenetics switching and sensing applications.

**Results Dissemination:** During the project, a number of papers were published or submitted with overall relevance to the research. The publications are listed below; see publication section for the detailed papers.

1. A key bidirectional switching issue in optogenetics emulated with laser dyes to illustrate its mitigation using nonlinear tools, Y. Wang, A. Goun, F. Laforge, Z. Quine, and H. Rabitz, Appl. Phys. Lett., DOI: 10.1063/5.0035183, (2021)

2. Coherent Control of Optogenetic Switching by Stimulated Depletion Quenching, Z. Quine, A. Goun, K. Gerhardt, J. Tabor, and H. Rabitz – arXiv:1810.11432v2

3. Analytical formulation of nonlinear optical responses in frequency comb spectroscopy, A. Chattopadhyay, A. Goun, D. Bondar, and H. Rabitz – in preparation

4. Discrimination of linear spectrally overlapping quantum systems with nonlinear frequency comb spectroscopy, A. Chattopadhyay, A. Goun, D. Bondar, and H. Rabitz – in preparation

5. Dual coherent and incoherent two-photon luminescence in single gold nanorods revealed by polarization and time resolved non-linear autocorrelation, D. Xie, F. Laforge, I. Grigorenko, and H. Rabitz, J. Optical Society of America B, 36, 1931-1936, DOI: 10.1364/JOSAB.36.001931, (2019)

6. Combining the synergistic control capabilities of modeling and experiments: Illustration of finding a minimum-time quantum objective, Q.-M. Chen, X. Yang, C. Arenz, R.-B. Wu, X. Peng, I. Pelczer, and H. Rabitz, Phys. Rev. A, 101, 032313, DOI:10.1103/PhysRevA.101.032313, (2020)

7. Shaped incoherent light for control of kinetics: Optimization of up-conversion hues in phosphors, F. Laforge, M. Kirschner, and H. Rabitz, J. Chem. Phys., 149, 054201, DOI: 10.1063/1.5035077, (2018)

8. Gaining Mechanistic Insight with Control Pulse Shaping: Application to the Dissociative Ionization of CH2BrI, X. Xing, R. Rey-de-Castro, and H. Rabtiz, J. Phys. Chem. A, 121, 8632-8641, DOI: 10.1021/acs.jpca.7b08835, (2017)

9. Ultrafast Photofragmentation of Ln(hfac)3 with a Proposed Mechanism for forming High Mass Fluorinated Projects, J. Chen, X. Xing, R. Rey-de-Castro, and H. Rabitz, Scientific Reports, 10, 7066, DOI: 10.1038/s41598-020-64015-2, (2020)

10. Deprotonation of Phenol linked to a silicon dioxide surface using Adaptive Feedback Laser Control with a Heterodyne Detected Sum Frequency Generation Signal, A. Goun, E. Frederick, A. O. Er, S. Bernasek, H. Rabitz – in preparation

11. Chemically Sensitive Fluorescence Imaging of Colliding Microdroplets, Z. Quine, A. Goun, F. Laforge, H. Rabitz, C. Law – in preparation

12. Selective photo-excitation of molecules via stimulated Raman pre-excitation, Y. Wang, F. Laforge, A. Goun, H. Rabitz – in preparation

13. Sequential optical response suppression for chemical mixture characterization, A. Magann, G. McCaul, H. Rabitz, D. Bondar – submitted to Quantum (2021)

14. Suppression of the Spectral Cross Talk of Optogenetic Switching by Stimulated Depletion Quenching. Theoretical Analysis, Z. Quine, A. Goun, H. Rabitz, arXiv:1811.05088v1

15. From Pulses to Circuits and Back Again: A Quantum Optimal Control Perspective on Variational Quantum Algorithms, A. Magann, C. Arenz, M. Grace, T.-S. Ho, R. Kosut, J. McClean, H. Rabitz, and M. Sarovar, PRX Quantum, 2, 010101, DOI: 10.1103/PRXQuantum.2.010101, (2021)

16. Learning Control of Quantum Systems Using Frequency-Domain Optimization Algorithms, D. Dong, C.-C. Shu, X. Xing, H. Ma, Y. Guo, and H. Rabitz, IEEE-TCST, 29(4), 1791, DOI: 10.1109/TCST.2020.3018500, (2021)

17. Optimal control of coupled quantum systems based on the first-order Magnus expansion: Application to

multiple dipole-dipole-coupled molecular rotors, A. Ma, A. Magann, T.-S. Ho, and H. Rabitz, Phys. Rev. A, 102, 013115: DOI: 10.1103/PhysRevA.102.013115, (2020)

18. Learning-Based Quantum Robust Control: Algorithm, Applications, and Experiments, D. Dong, X. Xing, H. Ma, C. Chen, Z. Liu, and H. Rabitz, IEEE Transactions on Cybernetics, 50, 3581-3593 DOI: 10.1109/TCYB. 2019.2921424, (2020)

19. Quantum optimal control of multiple weakly interacting molecular rotors in the time-dependent Hartree approximation, A. Magann, L. Chen, T.-S. Ho, and H. Rabitz, J. Chem. Phys., 150, 164303, DOI: 10.1063/1. 5091520, (2019)

20. Inherently trap-free convex for fully quantum optimal control, R.-B. Wu, Q. Sun, T.-S. Ho, and H. Rabitz, J. Math. Chem., 57, 2154-2167, DOI: 10.1007/s10910-019-01059-4, (2019)

21. Assessing the structure of classical molecular optimal control landscapes, C. Joe-Wong, T.-S. Ho, and H. Rabitz, Chem. Phys., 527, 110504, DOI: 10.1016/j.chemphys.2019.110504. (2019)

22. Singularity-free quantum tracking control of molecular rotor orientation, A. Magann, T.-S. Ho, and H. Rabitz, Phys. Rev. A, 98, 043429, DOI: 10.1103/PhysRevA.98.043429, (2018)

23. Reply to comment on “Control landscapes are almost always trap free: a geometric assessment”, B. Russell, R. -B. Wu, and H. Rabitz, J. Phys. A.: Math. Theor., 51, 508002, DOI: 10.1088/1751-8121/aaecf2, (2018)

24. Sampled-data design for robust control of open two-level quantum systems with operator errors, D. Dong, I. Petersen, Y. Wang, X. Yi, and H. Rabitz, IET Control Theory Appl., DOI: 10.1049/iet-cta.2016.0368, (2016)

25. Dependence of the quantum speed limit on system size and control complexity, J. Lee, C. Arenz, H. Rabitz, and B. Russell, New J. Phys, 20, 063002, DOI: 10.1088/1367-2630/aac6f3, (2018)

26. Data-driven gradient algorithm for high-precision quantum control, R.-B. Wu, B. Chu, D. Owens, and H. Rabitz, Phys. Rev. A, 97, 042122, DOI: 10.1103/PhysRevA.97.042122, (2018)

27. Controlling Qubit Networks in Polynominal Time, C. Arenz and H. Rabitz, Phys. Rev. Lett., 120, 220503, DOI: 10.1103/PhysRevLett.120.220503, (2018)

28. Control landscapes for a class of non-linear dynamical systems: sufficient conditions for the absence of traps, B. Russell, S. Vuglar, and H. Rabitz, J. Phys. A: Math. Theor., 51, 335103, DOI: 10.1088/1751-8121/aacc85, (2018)

29. The roles of drift and control field constraints upon quantum control speed limits, C. Arenz, B. Russell, D. Burgarth, and H. Rabitz, New J. Phys, 19, 103015, DOI: 10.1088/1367-2630/aa8242, (2017)

30. Searching for an optimal control in the presence of saddles on the quantum-mechanical observable landscape, G. Riviello, R.-B. Wu, Q. Sun, and H. Rabitz, Phys. Rev. A, 95, 063418, DOI: 10.1103/PhysRevA.95.063418, (2017)

31. How to Make Distinct Dynamical Systems Appear Spectrally Identical, A. Campos, D. Bondar, R. Cabrera, and H. Rabitz, Phys. Rev. Lett., 118, 083201, DOI: 10.1103/PhysRevLett.118.083201, (2017)

32. Control landscapes are almost always trap free: a geometric assessment, B. Russell, H. Rabitz, and R.-B. Wu, J. Phys. A: Math. Theor., 50, 205302, DOI: 10.1088/1751-8121/aa6b77, (2017)

33. Analytic Solutions to Coherent Control of the Dirac Equation, A. Campos, R. Cabrera, H. Rabitz, and D. Bondar, Phys. Rev. Lett., 119, 173203, DOI: 10.1103/PhysRevLett.119.173203, (2017)

34. Theory of molecular conductance using a modular approach, L.-Y. Hsu and H. Rabitz, J. Chem. Phys., 145, 234702, DOI: 10.1063/1.4972131, (2016)

35. Conductance and Activation Energy for Electron Transport in Series and Parallel Intramolecular Circuits, L.-Y.

Hsu, N. Wu, and H. Rabitz, Phys. Chem. Chem. Phys., 18, 32087-32095, DOI: 10.1039/C6CP06362D, (2016)

36. Exact-exchange optimized effective potential and memory effect in time-dependent density functional theory, S.-L. Liao, T.-S. Ho, H. Rabitz, and S.-I Chu, Eur. Phys. J. B, 91, 147, DOI: 10.1140/epjb/e2018-90084-x, (2018)

37. Peak Annotation and Verification Engine for Untargeted LC-MS Metabolomics, L. Wang, X. Xing, L. Chen, L. Yang, X. Su, H. Rabitz, W. Lu, and J. Rabinowitz, Anal. Chem., 91, 1838-1846, DOI: 10.1021/acs.analchem.8b03132, (2019)

38. Advances in Laser Chemistry in the Early Era, 1960-1985: Roots of Modern Quantum Control, J. Lee, F. Laforge, K. Kompa, H. Rabitz – in preparation for JCPL Perspective

**Honors and Awards:** Adjunct Professor, Swinburne University (Melbourne, Australia), February 2018 – February 2019

Visiting Scholar, University of New South Wales, 2019 - 2021

**Protocol Activity Status:**

**Technology Transfer:** Two unusual technology transfers occurred in this project linked to the fundamental principles of control.

1. Functional peptide discovery using amino acids as controls. The research considered optical fields as controls, but the same concepts were more broadly applied to chemical moieties considered as controls when bonded to a molecular scaffold. In recognition of this principle, using analogous theoretical tools as those utilized in this research, we were able to show that peptides may be effectively discovered as therapeutics drawing on special algorithms to guide their synthesis to have particular amino acid sequences, and thereby identify the entire peptide “control” landscape using advanced mathematical tools. In this case, the controls in the peptides are the discrete amino acids bonded to the underlying backbone. As a result, the company, Manzara Therapeutics, was formed and Princeton University has exclusively licensed this technology to the company. The fact that control using lasers or molecular fragments (i.e., amino acids) may be drawn together by the same concepts speaks to a larger control principle relevant in a wide-range of sciences. The technology transfer at hand is a clear example of a high value outcome in one direction (peptide discovery as pharmaceuticals) arising from research in a seemingly unrelated direction (optical laser pulse shape discovery for quantum control). This technology transfer is a quintessential example of surprises that can arise from basic research. Manzara Therapeutics commenced operations in the Fall of 2018 and several investors have provided initial funding of ~$1M. We are in the process of assembling a second round of funding. This funding is being utilized to demonstrate the scientific principle discussed above, while working with a family of synthesized peptide molecules and assessing their target binding properties. Interestingly, this technology transfer also has fundamental scientific significance by linking the basic control principles involved throughout our research including that of laser control.

2. Effective control of chemical kinetics through pulse shaped broad bandwidth incoherent light. This technology concerns the construction of an apparatus which can flexibly create incoherent light pulses with arbitrary wave forms for control of chemical kinetics for applications in chemistry and non-linear optical materials. This technology was motivated by the fact that experiments utilizing ultrafast laser technology are an extremely expensive venture (i.e., although for a high value application in the area of biological switches and sensors, the cost and complexity of the laser is not an issue). The audience for the present technology is the chemical kinetics and solid-state sensor communities, which are currently mired in the use of old technology essentially corresponding to operation with a light bulb. Our new technology has all the features of laser pulse shaping except only operating with broad bandwidth incoherent light created by an inexpensive simple array of laser diodes. Importantly, the cost of the instrument is roughly a factor of 50 less than that of an ultrafast laser setup. We have demonstrated proof of capability for the instrument technology with control over the luminescence from a solid-state material. We have filed a patent for the new instrument with an initial extremely positive response from the Patent Office. The company, Intelligent Materials Solutions, has licensed the technology for a variety of high value materials applications.

**PARTICIPANTS:**

**Participant Type:** PD/PI
**Participant:** Herschel Rabitz
**Person Months Worked:** 6.00 **Funding Support:**
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**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Tak-San Ho
**Person Months Worked:** 15.00 **Funding Support:**
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**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Christian Arenz
**Person Months Worked:** 8.00 **Funding Support:**
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**Participant Type:** Graduate Student (research assistant)
**Participant:** Peyman Azodi
**Person Months Worked:** 12.00 **Funding Support:**
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**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Roberto Rey-de-Castro
**Person Months Worked:** 5.00 **Funding Support:**
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**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Francois Laforge
**Person Months Worked:** 15.00 **Funding Support:**
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**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Alexei Goun
**Person Months Worked:** 15.00 **Funding Support:**
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**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Genyuan Li

**Person Months Worked:** 13.00 **Funding Support:**
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**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Xin Xu
**Person Months Worked:** 15.00 **Funding Support:**
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**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Xi Xing
**Person Months Worked:** 6.00 **Funding Support:**
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**Participant Type:** Graduate Student (research assistant)
**Participant:** David Hocker
**Person Months Worked:** 6.00 **Funding Support:**
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National Academy Member: N

**Participant Type:** Graduate Student (research assistant)
**Participant:** Andre Campos
**Person Months Worked:** 15.00 **Funding Support:**
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**Participant Type:** Graduate Student (research assistant)
**Participant:** Ayan Chattopadhyay
**Person Months Worked:** 6.00 **Funding Support:**
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**Participant:** Shanon Vuglar
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**Abstract:** This study probes the correlation between the conductance of a molecular wire (the property of a whole system) and its constituent backbone units (modules). By using a tight-binding Hamiltonian combined with single-particle Green's functions, we develop an approach that enables an estimate of a conductance decay constant in terms of the Hamiltonians of molecular backbone units and the couplings between two nearest-neighbor units in the off-resonant tunneling regime. For demonstration, we examine several representative molecular systems in a framework of the H¨uckel model (the simplest atomistic-level model). The H¨uckel model can be reduced to a single-orbital-per-site formulation [A. Nitzan, Annu. Rev. Phys. Chem. 52, 681 (2001)], and each energy level in the single-orbital-per-site picture can b expressed in an explicit form including the synergistic effect of all molecular orbitals of a molecular backbone unit. Based on the proposed approach, we showthe correspondence between the co
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**Authors:** Daoyi Dong, Ian R. Petersen, Yuanlong Wang, Xuexi Yi, Herschel Rabitz
**Keywords:** Data design, quantum systems
**Abstract:** This study proposes a sampled-data design method for robust control of open two-level quantum systems with operator errors. The required control performance is characterised using the concept of a sliding mode domain related to fidelity, coherence or purity. The authors have designed a control law offline and then utilise it online for a two-level system subject to decoherence with operator errors in the system model. They analyse three cases of approximate amplitude damping decoherence, approximate phase damping decoherence and approximate depolarising decoherence. They design specific sampling periods for these cases that can guarantee the required control performance.
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Publication Location:
**Article Title:** Conductance and activation energy for electron transport in series and parallel intramolecular circuits
**Authors:** Liang-Yan Hsu, Ning Wu, Herschel Rabitz
**Keywords:** intramolecular circuits, electron transport
**Abstract:** We investigate electron transport through series and parallel intramolecular circuits in the framework of the multi-level Redfield theory. Based on the assumption of weak monomer-bath couplings, the simulations depict the length and temperature dependence in six types of intramolecular circuits. In the tunneling regime, we find that the intramolecular circuit rule Gparallel ¼ G1 þ G2 þ 2 ffiffiffiffiffiffiffiffiffiffiffiffiffiffiffiffiffiffi G1G2 p is only valid in the weak monomer coupling limit. In the thermally activated hopping regime, for circuits based on two different molecular units Ma and Mb with distinct activation energies Eact,a 4 Eact,b, the activation energies of Ma and Mb in series are nearly the same as Eact,a while those in parallel are nearly the same as Eact,b. This study gives a comprehensive description of electron transport through intramolecular circuits from tunneling to thermally activated hopping. We hope that this work can motivate additional studies to design intramolecular
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** RSC Adv.
Publication Identifier Type: DOI Publication Identifier: 10.1039/C6RA16158H
Volume: 6 Issue: 95 First Page #: 92962
Date Submitted: 1/30/17 12:00AM Date Published:
Publication Location:
**Article Title:** Sampling-based robust control in synchronizing collision with shaped laser pulses: an application in charge transfer for H
**Authors:** Wei Zhang, Daoyi Dong, Ian R. Petersen, Herschel A. Rabitz
**Keywords:** laser pulses, optimal control
**Abstract:** In laser-assisted collisions, a control field may fail if we cannot precisely synchronize the colliding particles and the laser pulse. In this paper, we show that laser pulses that are robust in this situation can be obtained by a sampling-based method to achieve a desired charge transfer probability with limited sensitivity to the arrival time of the laser pulses. The time-dependent wave packet method and an adaptive target scheme are used in optimal control calculations based on an adiabatic two-state model of a H + D+ collision system. Several samples with different pulse arrival times are selected to construct robust fields at two different collision energies and the validity of these fields are examined by tests with additional samples. Excellent performance was obtained with the robust fields in both cases.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** Phys. Rev. A
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevA.95.063418
Volume: 95 Issue: First Page #: 063418
Date Submitted: 8/25/17 12:00AM Date Published: 1/31/17 4:22AM
Publication Location:
**Article Title:** Searching for an optimal control in the presence of saddles on the quantum mechanical observable landscape
**Authors:** Gregory Riviello, Re-Bing Wu, Qiuyang Sun, Herschel Rabitz
**Keywords:** saddles, Optimal Control
**Abstract:** The broad success of theoretical and experimental quantum optimal control is intimately connected to the topology of the underlying control landscape. For several common quantum control goals, including the maximization of an observable expectation value, the landscape has been shown to lack local optima if three assumptions are satisfied: (i) the quantum system is controllable, (ii) the Jacobian of the map from the control field to the evolution operator is full-rank, and (iii) the control field is not constrained. In the case of the observable objective, this favorable analysis shows that the associated landscape also contains saddles, i.e., critical points that are not local suboptimal extrema. In this paper, we investigate whether the presence of these saddles affects the trajectories of gradient-based searches for an optimal control. We show through simulations that both the detailed topology of the control landscape and the parameters of the system Hamiltonian influence whether
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** J. Phys. A
Publication Identifier Type: DOI Publication Identifier: 10.1088/1751-8121/aa6b77
Volume: 50 Issue: First Page #: 205302
Date Submitted: 8/25/17 12:00AM Date Published: 4/25/17 3:28AM
Publication Location:
**Article Title:** Control Landscapes are Almost Always Trap Free: A Geometric Assessment
**Authors:** Benjamin Russell, Herschel Rabitz, Re-Bing Wu
**Keywords:** trap-free landscapes, quantum control
**Abstract:** A proof is presented that almost all closed, finite dimensional quantum systems have trap free (i.e., free from local optima) landscapes for a large and physically general class of circumstances. This result offers an explanation for why gradient- based methods succeed so frequently in quantum control. The role of singular controls is analyzed using geometric tools in the case of the control of tbe propagator, and thus in the case of observables as well. Singular controls have been implicated as a source of landscape traps. The conditions under which singular controls can introduce traps, and thus interrupt the progress of a control optimization, are discussed and a geometrical characterization of the issue is presented. It is shown that a control being singular is not sufficient to cause control optimization progress to halt, and sufficient conditions for a trap free landscape are presented. It is further shown that the local
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 5-Submitted
**Journal:** Phys. Rev. Lett.
Publication Identifier Type: Publication Identifier:
Volume: Issue: First Page #:
Date Submitted: 1/30/17 12:00AM Date Published: 1/30/17 6:33PM
Publication Location:
**Article Title:** Meeting a high-performance quantum technology primary objective in the presence of secondary objectives
**Authors:** David Hocker, Herschel Rabitz
**Keywords:** quantum technology
**Abstract:** Emerging quantum technologies offer promise for a host of important applications. While such technologies can require satisfying multiple objectives, typically one primary objective is singled out for high performance optimization. We demonstrate that upon seeking near-optimal performance of the primary objective, then all secondary objectives asymptotically converge to respective constant values. This inevitable circumstance implies that if the secondary objectives lie beyond specified thresholds, then significant system re-engineering can be required to meet collective quantum technology performance demands.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** J. Math Chem
Publication Identifier Type: DOI Publication Identifier: 10.1007/s10910-019-01059-4
Volume: Issue: First Page #:
Date Submitted: 8/29/19 12:00AM Date Published: 8/21/19 3:37AM
Publication Location:
**Article Title:** Inherently trap-free convex landscapes for fully quantum optimal control
**Authors:** Qiuyang Sun, Re-Bing Wu, Tak-San Ho, Herschel Rabitz
**Keywords:** quantum control landscapes
**Abstract:** We present a comprehensive analysis of the landscape for fully quantum control landscape associated with the expectation value of an arbitrary observable of one quantum system controlled by another quantum system. It is shown that such quantum-quantum (Q-Q) control landscapes are convex, and hence devoid of local suboptima and saddle points that may exist in landscapes for quantum systems controlled by time-dependent classical fields. There is no controllability requirement for the Q-Q landscape to be trap-free, although the forms of Hamiltonians, the flexibility in choosing initial state of the controller, as well as the control duration, can influence the reachable optimal value on the landscape. All level sets of the Q-Q control landscape are connected convex sets. Finally, we show that the optimal solution of the Q-Q control landscape can be readily determined numerically, which is demonstrated using the Jaynes-Cummings model depicting a two-level atom interacting with a quantized
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** Phys. Rev. A
Publication Identifier Type: DOI Publication Identifier: 10.1021/acs.jpca.7b08835
Volume: 121 Issue: First Page #: 8632
Date Submitted: 8/8/18 12:00AM Date Published: 1/31/17 4:55AM
Publication Location:
**Article Title:** Gaining Mechanistic Insight with Control Pulse Slicing: Application to the Dissociative Ionization of CH2BrI
**Authors:** Xi Xing, Roberto Rey-de-Castro, Herschel Rabitz
**Keywords:** Control Pulse Slicing
**Abstract:** In quantum control experiments with shaped electromagnetic pulses, adaptive feedback control is often used to identify pulse shapes that can optimally steer the quantum system towards the desired outcome. However, gaining mechanistic information can pose a challenge due to the varied structural features of the control pulses and/or the often complex nature of the associated simulations of the experiments. In this article, we introduce Control Pulse Slicing (CPS) as an easy-to-implement experimental analysis tool that can be used directly in the laboratory without the need for modeling, to gain mechanistic insights in control experiments, regardless of whether the pulse is optimal or chosen by other means. As an illustration, we apply CPS to dissociative ionization of CH2BrI with mass spectral detection, where two pulses with similar intensities are investigated, with each capable of distinctively controlling the ratio of Br+/CH2Br+.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 5-Submitted
**Journal:** Automatica
Publication Identifier Type: Publication Identifier:
Volume: Issue: First Page #:
Date Submitted: 1/30/17 12:00AM Date Published: 1/30/17 6:58PM
Publication Location:
**Article Title:** REGULARITY AND GLOBALIZATION OF NONSMOOTH NEWTON METHODS FOR OPTIMAL CONTROL PROBLEMS OF DIFFERENTIAL ALGEBRAIC EQUATIONS
**Authors:** Jinhai Chen, Herschel Rabitz
**Keywords:** optimal control problems
**Abstract:** This paper focuses on regularity conditions and globalization techniques of non- smooth Newton methods of optimal control problems governed by mixed control-state constraints with differential algebraic equations. In contrast to the previous results, we analyze the lifting operator involved in the nonsmooth Newton methods, and establish the corresponding convergence results. We also give sufficient conditions for regularity of the generalized derivatives of the underlying system of nonsmooth operator equations. Moveover, combining with the constructed lifting operator, we develop two global smoothing gradient-like methods for the system of nonsmooth operator equations, and show that these gradient-like methods have global and local superlinear convergence under mild settings. Our numerical results are reported to support the efficiency of our global techniques.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** Physical Review Letters
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevLett.118.083201
Volume: 118 Issue: 8 First Page #: 083201
Date Submitted: 8/25/17 12:00AM Date Published: 2/24/17 5:00AM
Publication Location:
**Article Title:** How to Make Distinct Dynamical Systems Appear Spectrally Identical
**Authors:** Andre G. Campos, Denys I. Bondar, Renan Cabrera, Herschel A. Rabitz
**Keywords:** quantum, control
**Abstract:** We show that a laser pulse can always be found that induces a desired optical response from an arbitrary dynamical system. As illustrations, driving fields are computed to induce the same optical response from a variety of distinct systems (open and closed, quantum and classical). As a result, the observed induced dipolar spectra without detailed information on the driving field are not sufficient to characterize atomic and molecular systems. The formulation may also be applied to design materials with specified optical characteristics. These findings reveal unexplored flexibilities of nonlinear optics.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** New J. Phys
Publication Identifier Type: DOI Publication Identifier: 10.1088/1367-2630/aa8242
Volume: 19 Issue: First Page #: 103015
Date Submitted: 8/8/18 12:00AM Date Published: 10/8/17 5:19PM
Publication Location:
**Article Title:** The roles of drift and control field constraints upon quantum control speed limits
**Authors:** C. Arenz, B. Russell, D. Burgarth, and H. Rabitz
**Keywords:** speed limits, quantum control, unitary gates, quantum information
**Abstract:** In this work we derive a lower bound for the minimumtime required to implement a target unitary transformation through a classical time-dependent field in a closed quantum system. The bound depends on the target gate, the strength of the internal Hamiltonian and the highest permitted control field amplitude. These findings reveal some properties of the reachable set of operations, explicitly analyzed for a single qubit.Moreover, for fully controllable systems,we identify a lower bound for the time at which all unitary gates become reachable. Weuse numerical gate optimization in order to study the tightness of the obtained bounds. It is shown that in the single qubit case our analytical findings describe the relationship between the highest control field amplitude and the minimum evolution timeremarkably well. Finally, we discuss both challenges and ways forward for obtaining tighter bounds for higher dimensional systems, offering a discussion about the mathematical form and the physical
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** Phys. Rev. Lett.
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevLett.119.173203
Volume: 119 Issue: First Page #: 173203
Date Submitted: 8/8/18 12:00AM Date Published: 10/27/17 5:25PM
Publication Location:
**Article Title:** Analytic Solutions to Coherent Control of the Dirac Equation
**Authors:** A. Campos, R. Cabrera, H. Rabitz, and D. Bondar
**Keywords:** coherent control
**Abstract:** A simple framework for Dirac spinors is developed that parametrizes admissible quantum dynamics and also analytically constructs electromagnetic fields, obeying Maxwell's equations, which yield a desired evolution. In particular, we show how to achieve dispersionless rotation and translation of wave packets. Additionally, this formalism can handle control interactions beyond electromagnetic. This work reveals unexpected flexibility of the Dirac equation for control applications, which may open new prospects for quantum technologies.
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**Journal:** New J. Phys
Publication Identifier Type: DOI Publication Identifier: 10.1088/1367-2630/aac6f3
Volume: 20 Issue: First Page #: 063002
Date Submitted: 8/8/18 12:00AM Date Published: 6/5/18 5:40PM
Publication Location:
**Article Title:** Dependence of the quantum speed limit on system size and control complexity
**Authors:** J. Lee, C. Arenz, H. Rabitz, and B. Russell
**Keywords:** quantum control, quantum speed limit, quantum computation
**Abstract:** We extend the work in 2017 New J. Phys. 19 103015 by deriving a lower bound for the minimum time necessary to implement a unitary transformation on a generic, closed quantum system with an arbitrary number of classical control fields. This bound is explicitly analyzed for a specific N-level system similar to those used to represent simple models of an atom, or the first excitation sector of a Heisenberg spin chain, both of which are of interest in quantum control for quantum computation. Specifically, it is shown that the resultant bound depends on the dimension of the system, and on the number of controls used to implement a specific target unitary operation. The value of the bound determined numerically, and an estimate of the true minimum gate time are systematically compared for a range of system dimension and number of controls; special attention is drawn to the relationship between these two variables.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** J. Phys. Chem
Publication Identifier Type: DOI Publication Identifier: 10.1063/1.5035077
Volume: 149 Issue: First Page #: 054201
Date Submitted: 8/8/18 12:00AM Date Published: 8/8/18 1:50AM
Publication Location:
**Article Title:** Shaped Incoherent Light for Control of Chemical Kinetics: optimization of up-conversion hues in phosphors
**Authors:** F. Laforge, M. Kirschner, and H. Rabitz
**Keywords:** control
**Abstract:** We propose a method for interactively controlling multi-species atomic and molecular systems with incoherent light. The technique is referred to as shaped incoherent light for control (SILC), which entails dynamically tailoring the spectrum of a broadband incoherent source to control atomic and molecular scale kinetics. Optimal SILC light patterns can be discovered with adaptive learning techniques where the system's observed response is fed back to the control for adjustment aiming to improve the objective. To demonstrate this concept, we optimized a SILC source to optimally control the evolving hue in near-IR to visible upconverting phosphors, which share many similarities with chemical reaction kinetics including non-linear behavior. Thus, the results suggest that SILC may be a valuable tool for the control of chemical kinetics with tailored incoherent light
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**Journal:** J. Phys. A
Publication Identifier Type: DOI Publication Identifier: 10.1088/1751-8121/aacc85
Volume: 51 Issue: First Page #: 335103
Date Submitted: 8/8/18 12:00AM Date Published: 7/13/18 9:54PM
Publication Location:
**Article Title:** Control landscapes for a class of non-linear dynamical systems: sufficient conditions for the absence of traps
**Authors:** B. Russell, S. Vuglar, and H. Rabitz
**Keywords:** control theory, control landscapes, optimization
**Abstract:** We establish three tractable, jointly sufficient conditions for the control landscapes of non-linear dynamical systems to be trap free. Trap free landscapes ensure that local optimization methods (such as gradient ascent) can achieve monotonic convergence to the objective in both simulations and in practical circumstances. These results extend prior research primarily regarding the Schrödinger equation to a broader class of non-linear control problems encompassing both quantum and other dynamical systems. This outcome elucidates that these previous conclusions on quantum control landscapes were not specifically contingent upon any features unique to quantum dynamics. As an illustration of the new general results we demonstrate that they encompass end-point objectives for a general class of nonlinear control systems having the form of a linear time invariant term with an additional state dependent non-linear term.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 5-Submitted
**Journal:** J. Analytical Chemistry
Publication Identifier Type: Publication Identifier:
Volume: Issue: First Page #:
Date Submitted: 8/8/18 12:00AM Date Published:
Publication Location:
**Article Title:** Global parent ion annotation and formula assignment for untargeted LC-MS metabolomics
**Authors:** L. Wang, X. Xing, L. Chen, L. Yang, X. Su, H. Rabitz, W. Lu, and J. Rabinowitz
**Keywords:** metabolomics
**Abstract:** Untargeted metabolomics analysis can detect more than 10,000 peaks in a single LC-MS run. Which of these peaks correspond to metabolite molecular ions (“parent ions”) remains unclear. Here we introduce a Parent ion Annotation and Verification Engine (PAVE) for systematically annotating parent ions in untargeted metabolomics data. The workflow involves growing cells in 13C and 15N isotope-labeled media to identify biological peaks and their carbon and nitrogen atom counts. The labeling data are used to facilitate robust computational de-isotoping and deadducting. In addition, they enable filtering out of peaks for which the C atom counts are inappropriately low relative to the mass. The remaining peaks include both parent ions and fragments. To distinguish these, PAVE experimentally measures the response of each peak to a weak in-source collision-induced-dissociation voltage, which increases the signal for fragments while decreasing it for parent ions.
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**Journal:** Phys. Rev. A
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevA.97.042122
Volume: 97 Issue: First Page #: 042122
Date Submitted: 8/8/18 12:00AM Date Published: 4/24/18 6:02PM
Publication Location:
**Article Title:** Data-driven gradient algorithm for high-precision quantum control
**Authors:** R.-B. Wu, B. Chu, D. Owens, and H. Rabitz
**Keywords:** optimal control
**Abstract:** In the quest to achieve scalable quantum information processing technologies, gradient-based optimal control algorithms (e.g., GRAPE) are broadly used for implementing high-precision quantum gates, but their performance is often hindered by deterministic or random errors in the system model and the control electronics. In this paper, we show that GRAPE can be taught to be more effective by jointly learning from the design model and the experimental data obtained from process tomography. The resulting data-driven gradient optimization algorithm(d-GRAPE) can in principle correct all deterministic gate errors, with a mild efficiency loss. The d-GRAPE algorithm may become more powerful with broadband controls that involve a large number of control parameters, while other algorithms usually slow down due to the increased size of the search space. These advantages are demonstrated by simulating the implementation of a two-qubit controlled-NOT gate.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 0-Other
**Journal:** to be published
Publication Identifier Type: Publication Identifier:
Volume: Issue: First Page #:
Date Submitted: 10/2/19 12:00AM Date Published:
Publication Location:
**Article Title:** Coherent Control of Optogenetic Switching by Stimulated Depletion Quenching. Experimental Demonstrations
**Authors:** Z. Quine, A. Goun, K. Gerhardt, J. Tabor, H. Rabitz
**Keywords:** Optogenetics
**Abstract:** Optogenetics is a revolutionary new field of biotechnology, achieving optical control over biological functions in living cells by genetically inserting light sensitive proteins into cellular signaling pathways. Applications of optogenetic switches are expanding rapidly, but the technique is hampered by spectral cross-talk: the broad absorption spectra of compatible biochemical chromophores limits the number of switches that can be independently controlled and restricts the dynamic range of each switch. In the present work, we develop and implement a non-linear optical photoswitching capability, Stimulated Depletion Quenching (SDQ), is used to overcome spectral cross-talk by exploiting the molecules' unique dynamic response to ultrashort laser pulses. SDQ is employed to enhance the control of Cph8, a photo-reversible phytochrome based optogenetic switch designed to control gene expression in E. Coli bacteria. The Cph8 switch cannot be fully converted to its biologically inactive state
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 5-Submitted
**Journal:** ICVTS (Interactive CardioVascular and Thoracic Surgery)
Publication Identifier Type: Publication Identifier:
Volume: Issue: First Page #:
Date Submitted: 10/2/19 12:00AM Date Published:
Publication Location:
**Article Title:** Femtosecond removal of cardiac tissue along with Optical Coherence Tomography and Mass Spectroscopic assessment
**Authors:** A. Goun, H. Rabitz, N. Vasilyev
**Keywords:** Optical Coherence Tomography, Mass Spectroscopy
**Abstract:** The capability for removing cardiac tissue with high precision is demonstrated based on the use of a femtosecond laser combined with Optical Coherent Tomography (OCT) imaging and Mass Spectroscopy (MS) tissue analysis. Utilizing ex-vivo samples we demonstrate the spatial accuracy of femtosecond laser tissue removal with OCT. MS data is shown to be tissue specific and can potentially serve for real time tissue identification.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 5-Submitted
**Journal:** Risk Analysis
Publication Identifier Type: Publication Identifier:
Volume: Issue: First Page #:
Date Submitted: 10/2/19 12:00AM Date Published:
Publication Location:
**Article Title:** Global Sensitivity Analysis with Mixtures: A Generalized Functional ANOVA approach
**Authors:** E. Borgonovo, G. Li, J. Barr, E. Plischke, H. Rabitz
**Keywords:** ANOVA
**Abstract:** This work investigates the global sensitivity analysis of computer codes when analysts have available alternative plausible distributions for the model inputs. Analysts may decide to explore results under each distribution or to aggregate the distributions, assigning for instance a mixture. In the first case, we lose uniqueness of the sensitivity measures, in the second we lose independence. We analyze the methodological aspects that emerge from dealing with multiple distributions offering results for the properties of the terms of the functional ANOVA expansion and for the variance decomposition as well. We show that an approach based on the generalized functional ANOVA expansion is, in theory, successful in the presence of a mixture distribution. Numerically, we base the construction of the generalized function ANOVA effects on the D-MORPH regression. The resulting approach is compared against alternatives previously used in the literature. Our application addresses the calculation
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**Journal:** IEEE Transactions on Control Systems Technology
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Volume: 29 Issue: 4 First Page #: 1791
Date Submitted: 9/28/21 12:00AM Date Published: 6/10/21 4:00AM
Publication Location:
**Article Title:** Learning control of quantum systems using frequency-domain optimization algorithms
**Authors:** D. Dong, C.-C. Shu, J. Chen, X. Xing, H. Ma, Y. Guo, H. Rabitz
**Keywords:** femtosecond laser, frequency-domain optimization, learning control, quantum control
**Abstract:** We investigate two classes of quantum control problems by using frequency-domain optimization algorithms in the context of ultrafast laser control of quantum systems. In the first class of problems, the system model is known and a frequency-domain gradient-based optimization algorithm is applied for searching an optimal control field to selectively and robustly manipulate the population transfer in atomic rubidium. The other class of quantum control problems involves an experimental system with an unknown model. In this case, we introduce a differential evolution algorithm with a mixed strategy to search for optimal control fields and demonstrate the capability in an ultrafast laser control experiment for the fragmentation of Pr(hfac)3 molecules.
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 0-Other
**Journal:** arXiv
Publication Identifier Type: Publication Identifier:
Volume: Issue: First Page #:
Date Submitted: 10/2/19 12:00AM Date Published:
Publication Location:
**Article Title:** Quantum Control Landscapes Beyond the Dipole Approximation
**Authors:** B. Russell, H. Rabitz, R.-B. Wu
**Keywords:** quantum control landscapes
**Abstract:** We investigate the control landscapes of closed, n level quantum systems beyond the dipole approximation by including a polarization term in the Hamiltonian. Theoretical analysis of singular controls is presented, which are candidates for producing landscape traps; the results for singular controls are compared to their counterparts in the dipole approximation. A numerical analysis of the existence of traps in control landscapes beyond the dipole approximation is made in the n = 4 level case. A numerical exploration of these control landscapes is achieved by generating many random Hamiltonians which include a term quadratic in a single control field. The landscapes of such systems are found to be trap free in these studies. This result extends a great body of recent work on typical landscapes of quantum systems where the dipole approximation is made. We further investigate the relationship between the magnitude of the polarizability and the magnitude of the controls resulting from opt
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** The Journal of Chemical Physics
Publication Identifier Type: DOI Publication Identifier: 10.1063/1.5091520
Volume: 150 Issue: 16 First Page #: 164303
Date Submitted: 8/29/19 12:00AM Date Published: 4/1/19 4:00AM
Publication Location:
**Article Title:** Quantum optimal control of multiple weakly interacting molecular rotors in the time-dependent Hartree approximation
**Authors:** Alicia Magann, Linhan Chen, Tak-San Ho, Herschel Rabitz
**Keywords:** Quantum control, Hartree approximation
**Abstract:** We perform quantum optimal control simulations, based on the Time-Dependent Hartree (TDH) approximation, for systems of three to five dipole-dipole coupled OCS rotors. A control electric field is used to steer all of the individual rotors, arranged in chains and regular polygons in a plane, toward either identical or unique objectives. The goal is to explore the utility of the TDH approximation to model the field-induced dynamics of multiple interacting rotors in the weak dipole-dipole coupling regime. A stochastic hill climbing approach is employed to seek an optimal control field that achieves the desired objectives at a specified target time. We first show that multiple rotors in chain and polygon geometries can be identically oriented in the same direction; these cases do not significantly depend on the presence of the dipole-dipole interaction. Additionally, in particular geometrical arrangements, we demonstrate that individual rotors can be uniquely manipulated toward different o
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**Publication Type:** Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
**Journal:** Journal of the Optical Society of America B
Publication Identifier Type: DOI Publication Identifier: 10.1364/JOSAB.36.001931
Volume: 36 Issue: 7 First Page #: 1931
Date Submitted: 8/29/19 12:00AM Date Published: 6/1/19 4:00AM
Publication Location:
**Article Title:** Dual coherent and incoherent two-photon luminescence in single gold nanorods revealed by polarization and time-resolved nonlinear autocorrelation
**Authors:** Dan Xie, François O. Laforge, Ilya Grigorenko, Herschel A. Rabitz
**Keywords:** coherent and incoherent luminescence
**Abstract:** It is well known that gold nanorods (AuNRs) readily emit two-photon luminescence (TPL) when excited by a broad bandwidth laser pulse that is tuned to the AuNRs' localized surface plasmon resonance. The nature of the mechanism (i.e., especially its degree of coherence) is under active debate. In this work, we measured the TPL emission from single nanorods while varying the angle ? between the linearly polarized laser electric field and the nanorod' s orientation. Data were best fit with a linear combination of cos4 ? and cos2 ? functions. While the former function may represent TPL signals arising from both coherent and incoherent processes, the later function is indicative of a purely incoherent process. To further validate this assessment, we measured TPL emission from single nanorods in a time-resolved collinear autocorrelation setup. The autocorrelation signal exhibited a large peak at zero delay, which is characteristic of coherent two-photon absorption and two lower intensity wing
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**Journal:** Physical Review A
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevA.98.043429
Volume: 98 Issue: 4 First Page #:
Date Submitted: 8/29/19 12:00AM Date Published: 10/1/18 4:00AM
Publication Location:
**Article Title:** Singularity-free quantum tracking control of molecular rotor orientation
**Authors:** Alicia Magann, Tak-San Ho, Herschel Rabitz
**Keywords:** Quantum control
**Abstract:** Quantum tracking control aims to identify applied fields to steer the expectation values of particular observables along desired paths in time. The associated temporal fields can be identified by inverting the underlying dynamical equations for the observables. However, fields found in this manner are often plagued by undesirable singularities. In this paper, we consider a planar molecular rotor and derive singularity-free tracking expressions for the fields that steer the expectation of the orientation of the rotor along the desired trajectories in time. Simulations are presented that utilize two orthogonal control electric fields to drive the orientation of the rotor along a series of designated tracks.
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**Journal:** The European Physical Journal B
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Volume: 91 Issue: 7 First Page #:
Date Submitted: 8/29/19 12:00AM Date Published: 7/1/18 4:00AM
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**Article Title:** Exact-exchange optimized effective potential and memory effect in time-dependent density functional theory
**Authors:** Sheng-Lun Liao, Tak-San Ho, Herschel Rabitz, Shih-I Chu
**Keywords:** Time-dependent density functional theory
**Abstract:** The memory effect in time-dependent density functional theory (TDDFT) is important in simulating many time-dependent physical processes, and its implementation in real time has been a longstanding challenge, thus limiting most of TDDFT applications to either adiabatic or linear-response regime. In this paper, we conduct the non-adiabatic calculations for a one-dimensional two-electron Helium model in a triplet state using the recently formulated Sturm-Liouville-type time-local equation for the time-dependent optimized effective potential (TDOEP) with the exact exchange functional, and the results agree with the exact time-dependent Schrodinger equation solutions. It is also found that the time-dependent dipole moment and probability density calculated from the TDOEP approach are more accurate than those from the adiabatic time-dependent Krieger-Li-Iafrate (TDKLI) approximation and the adiabatic local spin density approximation. Specifically, the non-adiabatic and memory-dependent terms
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**Journal:** Physical Review Letters
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Volume: 120 Issue: 22 First Page #:
Date Submitted: 8/29/19 12:00AM Date Published: 5/1/18 4:00AM
Publication Location:
**Article Title:** Controlling Qubit Networks in Polynomial Time
**Authors:** Christian Arenz, Herschel Rabitz
**Keywords:** Qubit systems
**Abstract:** Future quantum devices often rely on favorable scaling with respect to the number of system components. To achieve desirable scaling, it is therefore crucial to implement unitary transformations in a time that scales at most polynomial in the number of qubits. We develop an upper bound for the minimum time required to implement a unitary transformation on a generic qubit network in which each of the qubits is subject to local time dependent controls. Based on the developed upper bound, the set of gates is characterized that can be implemented polynomially in time. Furthermore, we show how qubit systems can be concatenated through controllable two body interactions, making it possible to implement the gate set efficiently on the combined system. Finally, a system is identified for which the gate set can be implemented with fewer controls.
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**Journal:** Appl. Phys. Lett.
Publication Identifier Type: DOI Publication Identifier: 10.1063/5.0035183
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**Article Title:** A key bidirectional switching issue in optogenetics emulated with laser dyes to illustrate its mitigation using nonlinear optical tools
**Authors:** Y. Wang, A. Goun, F. Laforge, Z Quine, and H. Rabitz
**Keywords:** optogenetic switching
**Abstract:** Conventionally, bidirectional optogenetic switches are controlled with linear excitation (e.g., monochromatic CW light), which has serious limitations when there is a spectral overlap between the “on” and “off” states of the switch. The spectral crosstalk lowers the selectivity of the photoactivation step, resulting in a moderate photoequilibrium value and a poor dynamic range for the switch. Using stimulated depletion quenching (SDQ), which is a nonlinear optical strategy similar to STED, we demonstrate enhanced photoactivation selectivity on one side of the switch, thus shifting the photoequilibrium beyond what is achievable with CW light. The discussion is built around Cph8 as a prototypical bidirectional optogenetic switch lacking complete photoreversibility upon CW excitation because of the spectral crosstalk. We use two fluorescent dyes as analogues to emulate the key spectral feature of the on and off states of Cph8; in this way, we focus on the initial photoactivation step and
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**Journal:** In Preparation
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**Article Title:** Analytical formulation of nonlinear optical responses in frequency comb spectroscopy
**Authors:** A. Chattopadhyay, A. Goun, D. Bondar, and H. Rabitz
**Keywords:** Nonlinear Spectroscopy, optical
**Abstract:** A broad class of nonlinear spectroscopies ranging from pump-probe experiments, photon-echoes to CARS (coherent anti-Stokes Raman spectroscopy) are well understood in terms of the nonlinear optical polarization due to a chemical species interacting with a laser field. However, these relatively weak nonlinear signals can be difficult to recover in the background of the much stronger laser excitation field. We theoretically investigate the use of frequency shifted optical frequency combs (OFCs) in efficiently detecting the 3A3-order nonlinear optical response due to a large chromophore modeled quantum mechanically, with the exciting fields being treated classically. Utilizing the fine comb structure and nonlinear optical mixing in molecular systems, the resultant polarization response signals are conveniently obtained as easily discernible spikes (comb lines) in the “field-free” region between consecutive comb lines. Invoking the dipole approximation and modeling the electric field of the
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**Article Title:** Discrimination of linear spectrally overlapping quantum systems with nonlinear frequency comb spectroscopy
**Authors:** A. Chattopadhyay, A. Goun, D. Bondar, and H. Rabitz
**Keywords:** quantum systems, spectroscopy
**Abstract:** Large polyatomic chromophore molecules are often hard to discriminate when present as a mixture, as they can have overlapping linear response (absorption) spectra. Nonlinear response functions carry additional spectroscopic information compared to their linear counterparts. As a result, systems with very similar or even identical linear responses can have differences in their nonlinear spectra. In this article we make use of nonlinear frequency comb-based spectroscopy to generate nonlinear response from a mixture of systems with similar linear responses. We theoretically describe a process of discriminating such systems utilizing spectroscopic advantages offered by the use of frequency-shifted optical combs. To this end we introduce an algorithm, using matrix decomposition techniques, for designing heterodyne fields that selectively detect the nonlinear response due to a given molecule over those due to other similar molecules in a mixture. The observed nonlinear responses due to simil
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**Article Title:** Chemically Sensitive Fluorescence Imaging of Colliding Microdroplets
**Authors:** Z. Quine, A. Goun, F. Laforge, H. Rabitz, C. Law
**Keywords:** Fluorescence
**Abstract:** We present a simple optical capability for generating spatially resolved chemical concentration maps of mixing fluids using a chemically sensitive dye, HPTS, detected by planar laser induced fluorescence (PLIF). To demonstrate an application of this capability we investigate the collision and mixing of a pair of micro-droplets in air. The two micro-droplets are composed of different fluids, methanol and water, with the dye initially in the methanol droplet. When the droplets collide the fluorescence of the dye shifts from blue to green as the solvent environment changes. A series of spectral-temporal images of the collision and subsequent mixing are recorded, from which we extract the distribution of the two intermixing droplet species reflected in the spatially resolved dye spectra. Images reveal material transfer between droplets in both coalescing and non-coalescing droplet collisions.
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**Journal:** In preparation
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**Article Title:** Selective photo-excitation of molecules via stimulated Raman pre-excitation
**Authors:** Y. Wang, F. Laforge, A. Goun, and H. Rabitz
**Keywords:** Spectroscopy, Raman Excitation
**Abstract:** Double resonance excitation, where the energies of vibrational and electronic molecular transitions are combined in a single, sequential excitation process, has been invented in the 1970s but has only been recently applied to microscopy thanks to the immense progress and ubiquity of use of Raman spectroscopy. The value of the technique is in combining the chemical selectivity of IR or Raman excitation with the much larger cross-sections of electronic transitions. Recently, it has been shown to be particularly suited for the detection and identification of chromophores at low concentrations and in the presence of spectral crosstalk. However, despite its low quantum yield per pulse sequence, we believe the technique has potential for selective photochemical transformations. There are some cases (e.g., the selective excitation of optogenetic switches) where the low yield may be overcome by repeated excitations to build up chemically relevant concentrations. Here we show that double resona
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**Journal:** arXiv
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**Article Title:** Suppression of the Spectral Cross Talk of Optogenetic Switching by Stimulated Depletion Quenching. Theoretical Analysis
**Authors:** Z. Quine, A. Goun, H. Rabitz
**Keywords:** Optogenetics
**Abstract:** Optogenetics is a rapidly growing field of biotechnology, potentially allowing a deeper understanding and control of complex biological networks. The major challenge is the multiplexed control of several optogenetic components in the presence of significant spectral cross talk. We propose and demonstrate through simulations a new control approach of Stimulated Depletion Quenching. This approach is applied to the phytochrome Cph8 bidirectional optogenetic switch, and the results show significant improvement of its dynamic range.
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**Article Title:** From Pulses to Circuits and Back Again: A Quantum Optimal Control Perspective on Variational Quantum Algorithms
**Authors:** Alicia B. Magann, Christian Arenz, Matthew D. Grace, Tak-San Ho, Robert L. Kosut, Jarrod R. McClean
**Keywords:** quantum algorithms, quantum technologies
**Abstract:** The last decade has witnessed remarkable progress in the development of quantum technologies. Although fault-tolerant devices likely remain years away, the noisy intermediate-scale quantum devices of today may be leveraged for other purposes. Leading candidates are variational quantum algorithms (VQAs), which have been developed for applications including chemistry, optimization, and machine learning, but whose implementations on quantum devices have yet to demonstrate improvements over classical capabilities. In this Perspective, we propose a variety of ways that the performance of VQAs could be informed by quantum optimal control theory. A major theme throughout is the need for sufficient control resources in VQA implementations; we discuss different ways this need can manifest, outline a variety of open questions, and look to the future.
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**Article Title:** Optimal control of coupled quantum systems based on the first-order Magnus expansion: Application to multiple dipole-dipole-coupled molecular rotors
**Authors:** Andrew Ma, Alicia B. Magann, Tak-San Ho, Herschel Rabitz
**Keywords:** coupled quantum systems, optimal control, Magnus expansion
**Abstract:** This paper presents a method for performing approximate optimal control simulations for quantum systems with multiple coupled degrees of freedom. The time evolution is simulated using the first-order Magnus expansion in the interaction picture, where the couplings between different degrees of freedom are treated as the perturbation. A numerical implementation procedure is presented that leverages upon pairwise couplings and the separability of the zeroth-order time evolution operator to achieve a reduced computational cost, which is analyzed with respect to the number of degrees of freedom. The formulation is compatible with gradient-free methods to optimize the control field, and a stochastic hill climbing algorithm is adopted for this purpose. As illustrations, optimal control simulations are performed for systems of two and three dipole-dipole-coupled molecular rotors under the influence of a control field. For the two-rotor system, the field is optimized to achieve either orientati
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**Article Title:** Reply to comment on ‘control landscapes are almost always trap free: a geometric assessment’
**Authors:** Benjamin Russell, Rebing Wu, Herschel Rabitz
**Keywords:** Control landscapes, quantum control, quantum systems
**Abstract:** This paper is a rebuttal of the comment by Zhdanov (2018 J. Phys. A: Math. Theor.) with regard to our paper (Russell et al 2017 J. Phys. A: Math. Theor. 50 205302). Parts of the comment either misrepresent the original claims in Russell et al (2017 J. Phys. A: Math. Theor. 50 205302) or consider issues that go beyond the scope of Russell et al (2017 J. Phys. A: Math. Theor. 50 205302). We address all of these points in the rebuttal. A remaining item about the prospect of an accumulation of null sets was already considered in a prior work (Russell et al 2018 J. Phys. A: Math. Theor. 51 335103). Importantly, the comment fails to consider the vast and continually growing set of successful quantum control tests under well-defined conditions, which serves as foundational evidence for the results in Russell et al (2017 J. Phys. A: Math. Theor. 50 205302). In summary, the remarks in the comment either misconstrue the claims of Russell et al (2017 J. Phys. A: Math. Theor. 50 205302) or raise p
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**Article Title:** Peak Annotation and Verification Engine for Untargeted LC–MS Metabolomics
**Authors:** Lin Wang, Xi Xing, Li Chen, Lifeng Yang, Xiaoyang Su, Herschel Rabitz, Wenyun Lu, Joshua D. Rabino
**Keywords:** mass spectrometry, adducts, metabolism, labeling, ions
**Abstract:** Untargeted metabolomics can detect more than 10,000 peask in a single LC-MS run. The correspondence between these peaks and metabolites, however, remains unclear. Here, we introduce a Peak Annotation and Verification Engine (PAVE) for annotating untargeted microbial metabolomics data. The work?ow involves growing cells in 13C and 15N isotope- labeled media to identify peaks from biological compounds and their carbon and nitrogen atom counts. Improved deisotoping and deadducting are enabled by algorithms that integrate positive mode, negative mode, and labeling data. To distinguish metabolites and their fragments, PAVE experimentally measures the response of each peak to weak in-source collision induced dissociation, which increases the peak intensity for fragments while decreasing it for their parent ions. The molecular formulas of the putative metabolites are then assigned based on database searching using both m/z and C/N atom counts. Application of this procedure to Saccharomyces ce
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**Article Title:** Advances in Laser Chemistry in the Early Era, 1960-1985: Roots of Modern Quantum Control
**Authors:** J. Lee, F. O. Laforge, K. L. Kompa, and H. A. Rabitz
**Keywords:** laser-selective chemistry, laser, quantum control
**Abstract:** Physicists revolutionized the scientific world when they invented the laser in 1960. The ground-breaking properties of the new light sources quickly captured the imagination of chemists who dreamed of `laser-selective chemistry', i.e., controlling chemical reactions with lasers. During the next two decades, fruitful interplay between theoretical and experimental progress in laser-selective chemistry kept the dream alive but generally did not lead to routine real-world applications begging the question: “what came out of this era?" In striving to achieve laser-selective chemistry scientists gradually realized the immense complexity of the problem of controlling quantum objects in general and, by doing so, they opened the doors to a new, broader scientific field of modern research called `quantum control'. Nowadays however, if we asked a student of quantum control about the origin of the field they would probably trace it to around 1985 which is when most reviews of quantum control start
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**Journal:** Quantum
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**Article Title:** Sequential optical response suppression for chemical mixture characterization
**Authors:** A. B. Magann, G. McCaul, H. A. Rabitz, and D. I. Bondar
**Keywords:** quantum tracking control, spectroscopy
**Abstract:** The characterization of mixtures of non-interacting, spectroscopically similar quantum components has important applications in chemistry, biology, and materials science. We introduce an approach based on quantum tracking control that allows for determining the relative concentrations of constituents in a quantum mixture, using a single pulse which enhances the distinguishability of components of the mixture and has a length that scales linearly with the number of mixture constituents. To illustrate the method, we consider two very distinct model systems: mixtures of diatomic molecules in the gas phase, as well as solid-state materials composed of a mixture of components. A set of numerical analyses are presented, showing strong performance in both settings.
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**Paper Title:** Optimal Dynamic Control of Multiple Quantum Systems for Multiplexed Bio-Network Analysis and Optogenetics
**Authors:** Herschel Rabitz, Alexei Goun
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**Paper Title:** Efficient Non-Linear Optical Spectroscopy with Multiple High-Frequency Modulated Optical Frequency Combs
**Authors:** A. Chattopadhyay, A. Goun, H. Rabitz
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**Partners**

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I certify that the information in the report is complete and accurate:
Signature: Herschel Rabitz
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Additional Information

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