Optimal Identification of Hamiltonian Information by Closed-Loop Laser Control of Quantum Systems

J. M. Geremia* and Herschel Rabitz†

Department of Chemistry, Princeton University, Princeton, New Jersey 08544

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A closed loop learning control concept is introduced for teaching lasers to manipulate quantum systems for the purpose of optimally identifying Hamiltonian information. The closed loop optimal identification algorithm operates by revealing the distribution of Hamiltonians consistent with the data. The control laser is guided to perform additional experiments, based on minimizing the dispersion of the distribution. Operation of such an apparatus is simulated for two model finite dimensional quantum systems.

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Detailed knowledge of quantum Hamiltonians is essential for a broad variety of applications. Various types of laboratory data have served as sources to extract the Hamiltonian information. Typically, experiments for this purpose are chosen based on practical considerations, as well as intuition regarding the significance of the data for the ultimate sought-after Hamiltonian. The relationship between the observations and the underlying Hamiltonian is generally highly nonlinear. Choices of which experiments to perform based on intuition alone may unwittingly provide false guidance. This Letter introduces a closed loop procedure for optimally identifying (OI) Hamiltonian information, taking advantage of the freedom inherent with shaped control fields [1] to manipulate the physical system.

Execution of the OI concept is particularly attractive in situations that may exploit the emerging capabilities of closed loop laser control of quantum dynamics phenomena [2,3]. Successful control over the dynamics of a quantum system generally relies on high finesse manipulation of constructive and destructive quantum wave interferences through suitably shaped laser fields [4]. Recognizing that the outcome of such experiments can be sensitive to the often imprecisely known Hamiltonian led to the suggestion of employing closed loop laboratory techniques for directly teaching lasers how to achieve quantum system control [2]. This procedure circumvents the need for prior knowledge about the system Hamiltonian, and a growing number of successful closed loop quantum control experiments have been reported [3,5–10]. This Letter shows that similar closed loop laboratory operations can be redirected for the purpose of extracting information about the underlying Hamiltonian, rather than meeting some particular observational target. As the goal is to ultimately obtain high quality Hamiltonian structure (e.g., matrix elements, potentials, dipoles, etc.), the effects of noise in the driving laser, as well as in the observations, need to be considered. Ameliorating this situation is the ability of the OI algorithm to guide the controlled collection of data, to reveal an optimal set of experiments that are robust to the noise and optimal for inversion. The goal is to have the OI machine deduce tailored controls and associated observations that leave an absolute minimum of uncertainty about the true underlying Hamiltonian.

Figure 1 presents an overall view of the basic OI machine components. The inversion algorithm in the loop extracts Hamiltonian information from the laboratory observations, while the learning control algorithm suggests new laser pulses guided by the goal of improving the quality of the inversion. It is essential that the inversion algorithm provide as much information as possible about the full distribution of Hamiltonians consistent with the

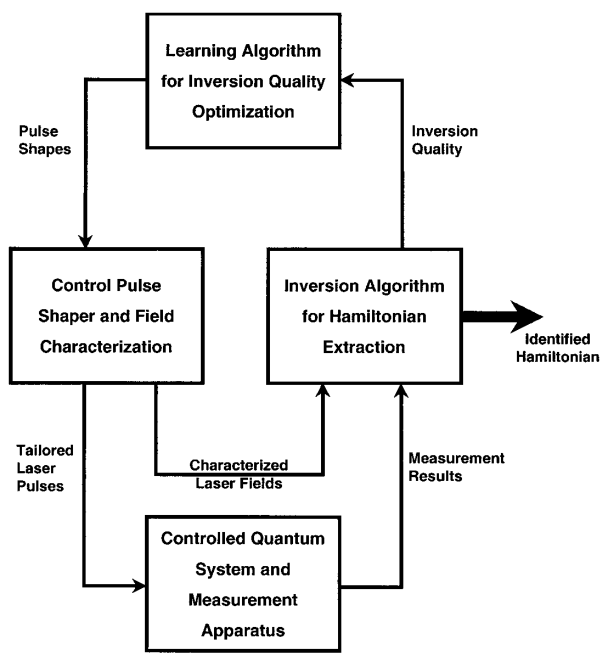

FIG. 1. The components of an apparatus operating under closed loop to optimally identify system Hamiltonians: The decision on which new control experiments to perform in the loop is based on the goal of attaining the best quality Hamiltonian information.

data in order for the learning control algorithm to make reliable choices for minimizing the breadth of the distribution. As with closed loop learning control aimed at a specific dynamical target [2,5–10], the learning control algorithm in the OI loop can attractively function using genetic algorithms (GA) [11–13]. Most importantly, the requirement that the inversion algorithm produce a distribution of solutions (i.e., Hamiltonians) points towards GA's as the procedure of choice in that component as well.

Considering the above issues, each excursion around the loop will involve a family of K laser control fields $\varepsilon_k(t)$, $k = 1, 2, \ldots, K$, where a particular field $\varepsilon_k(t)$ is associated with a concomitant laboratory observation Φ_k^{lab}. In practice, the action of each field upon the quantum system would be repeated a number of times due to field and observation noise, yielding a distribution of observations $P_k^{\text{lab}}[\Phi_k^{\text{lab}}, \varepsilon_k]$, which may not be Gaussian. In turn, the inversion of the kth data set would result in a distribution $q_k(H)$ of consistent Hamiltonians H. Here, H might be described by a collection of matrix elements or some other representation of the relevant Hamiltonian components. The shape of the distribution $q_k(H)$ is dictated by the laboratory data distribution and the various dynamical intricacies driving the phenomena. The GA guiding the inversion would best be performed with a very large population (e.g., $N_s \gtrsim 500$) of Hamiltonians H_s, $s = 1, 2, \ldots, N_s$ to generate the distribution $q_k(H)$. The inversion would be carried out aiming at a match between the laboratory and theoretical observational distributions, P_k^{lab} and P_k^{th}, where the inversion would be performed taking into account the reported errors in the control field and observations. The cost function guiding the GA for this purpose, $J_{\text{inv}}^k = \frac{1}{N_s}\sum_{s=1}^{N_s} J_{\text{inv}}(H_s, \Phi_k^{\text{lab}}, \varepsilon_k)$, $k = 1, 2, \ldots, K$ could have any of a variety of standard forms. A logical choice is to minimize the norm $\|P_k^{\text{lab}} - P_k^{\text{th}}\|$ over the set of consistent Hamiltonians $\{H_s\}$ to finally yield the distribution of acceptable Hamiltonians $q_k(H)$ associated with the kth control field. A suitable error metric ΔH_k may be obtained (e.g., the left and right relative error variance of the relevant components of H) from $q_k(H)$. Then, armed with the error metric $\Delta H_k > 0$, a cost function of the general form $J_{\text{cont}}^k = J_{\text{inv}}^k + \Delta H_k + \|\varepsilon_k\|$, $k = 1, 2, \ldots, K$ may be utilized to seek better control fields for another excursion around the loop. The norm $\|\varepsilon_k\|$ serves to maintain control field simplicity, build in any laboratory constraints on the field, or guide the apparatus away from introducing laser pulse shapes that have little significance for a successful inversion. Thus, the choice of a new family of control fields $\{\varepsilon_k(t)\}$ for another excursion around the loop would be made, balancing the inversion quality J_{inv}^k (which is expected to be readily achieved, such that $J_{\text{inv}}^k \simeq 0$) against minimization of the Hamiltonian uncertainty ΔH_k, along with some weighting on $\|\varepsilon_k\|$ to keep the control field physically reasonable. The new family of control fields $\{\varepsilon_k(t)\}$ would be deduced by the GA considering the performance of the prior family of controls in successfully minimizing J_{cont}^k. As with the current closed loop quantum control experiments, there is considerable flexibility in all of these algorithmic aspects, including in the choice of operating conditions for the two GA's guiding the inversion and choice of controls [2,5–13]. Although the OI machine performance [i.e., the production of an absolute minimum dispersion in the distribution $q_k(H)$] for at least one value of k can depend on these algorithmic details, the overall concept remains the same.

Closed loop operations for system identification have a precedent in the engineering disciplines [14], but there are special distinctions that arise in the present circumstances. Typically, in engineering, inversion is carried out for the purpose of learning a portion of a model to improve an ultimate system control gain. In the case of quantum mechanics, a more general desire is to seek the Hamiltonian for other ancillary purposes besides control. Second, most common engineering applications involve maintaining operational stability, which usually is expressed in terms of a locally linear system model. In contrast, in quantum mechanics, most applications will not be amenable to perturbation theory, thereby producing a nonlinear modeling and identification problem. Last, the ability to perform massive numbers of control laser experiments [3,4] opens up a special opportunity, difficult to achieve in engineering, where it is often prohibitive to thoroughly explore the analogous model distributions q_k.

Two illustrations are carried out to demonstrate the OI machine concepts in Fig. 1. Both illustrations involve a system corresponding to HF [15] having a Hamiltonian of the form $H = H_0 - \mu \cdot \varepsilon(t)$, where H_0 is field-free and μ is the system dipole. This system just serves as a model to test the OI algorithm with realistic Hamiltonian matrix elements. In practice, the iterative inversion always uses the relative error metric ΔH_k, which is the ratio of current estimated errors to the estimate for the associated matrix elements in H_k. Thus, the normalization places the small and large matrix elements on an equal footing and provides a rather generic test of the OI algorithm. The simulations are carried out with noise in the control fields and in the observations, where the overall error distribution $P_k^{\text{lab}}[\Phi_k^{\text{lab}}, \varepsilon_k]$ is assumed to arise from a root mean square combination of both error sources. The GA's used for inversion and control in both illustrations performed in a standard fashion with mutation and crossover operations [11–13]. For demonstration of the OI concept, the control fields were chosen to have the form $\varepsilon_k(t) = \exp[-(t/\sigma)^2]\sum_\ell a_\ell^k \cos(\omega_\ell + \phi_\ell^k)$, where the actual controls are the amplitudes a_ℓ^k and phases ϕ_ℓ^k associated with the system resonance frequencies ω_ℓ. The fields had a width of $\sigma = 200$ fs.

The first illustration consists of a ten-level Hamiltonian, with the data $\phi_{k,i}^{\text{lab}}$, $i = 1, 2, \ldots, 10$ for the kth control experiment being the state populations at the time $T = 1.5$ ps, after the control field $\varepsilon_k(t)$ evolved the system from its ground state. The matrix H_0 was taken as diagonal and known, with the goal of deducing the real dipole matrix elements $\{\mu_{nm}\}$. The latter elements [15] had the natural trend of having larger magnitude associated with smaller values of $|n - m|$, $n \neq m$. With $\mu_{nn} = 0$, there are a total of 45 unknown matrix elements to be determined by the closed loop OI operation. The inversion quality of the 17 most significant elements $\mu_{n,n\pm1}$, $\mu_{n,n\pm2}$ are reported here, although comparable high quality information was obtained for the entire matrix. The control fields guiding the machine operations in Fig. 1 were capable of inducing up to third order transitions $|n - m| \leq 3$. Figure 2 shows the error distribution for the extracted 17 matrix elements $\mu_{n,n\pm1}$, $\mu_{n,n\pm2}$, considering the laboratory error distributions $P_k^{\rm lab}$ as uniform with a standard deviation of 1%. Excellent quality dipole matrix elements were extracted after a number of cycles around the loop in Fig. 1. The mean error of the extracted elements was less than 0.1%, which is an order of magnitude smaller than the laboratory data error. Importantly, this high quality extracted information was obtained using a single optimally deduced control field and its ten observations. The fact that a successful inversion may be performed with only ten observations to determine 17 unknown matrix elements reflects the fact that the relation between the data and the sought-after matrix elements in the inversion is highly nonlinear. This nonlinear feature is advantageous in aiding the OI procedure to find a single optimal control experiment that produces a high quality Hamiltonian consistent with the data. The OI process required less than 500 experiments to deduce the single optimal experiment. As a point of comparison, a series of nonoptimal reference experiments were simulated, involving 500 randomly chosen amplitudes and phases for the control fields and, therefore, 5000 population observations. Utilizing all of this collective data for inversion produced the associated error distribution also shown in Fig. 2. Despite the fact of having an overwhelming amount of data (i.e., 5000 data points to determine 17 unknown matrix elements), the quality of the nonoptimal inversion is far inferior to that obtained by OI using a single identified field and its associated ten observations. To further illustrate the power of the OI machine concept, additional simulations were carried with observation errors of 2% and 5%. Despite these quite significant increases in laboratory error, the associated dipole matrix element error distributions were almost identical to the optimal one shown in Fig. 2, although the optimal fields depended on the amount of noise in the data. This behavior indicates that the OI machine sought out the best control field to filter out the data noise under each set of conditions.

As a second illustration, both H_0 (i.e., the potential portion of the field-free Hamiltonian) and μ were treated as unknowns in a corresponding real Hamiltonian matrix of dimension 8. Again, considering that $\mu_{nn} = 0$, there are a total of 64 matrix elements as unknowns in H_0 and μ. These matrix elements are represented in the basis associated with the observed populations. The data consisted of the populations sampled at evenly spaced times out to 1 ps, and the laboratory error was taken as 2%. Figure 3 shows the effect of increasing amounts of data upon the quality of the inversion of both H_0 and μ. With a set of eight observations at a single time of 1 ps, the average error for μ is already less than the laboratory error, while that of H_0 is somewhat larger. The situation improves when taking in data at a second time of 0.5 ps, and finally, at four times spaced by 0.25 ps, for a total of 32 data points. The quality of both the extracted H_0 and μ matrix elements is excellent, with average errors being at least an order of magnitude smaller than the laboratory data error. In contrast, Fig. 3 also shows that taking a total of 200 data points using a set of random control field

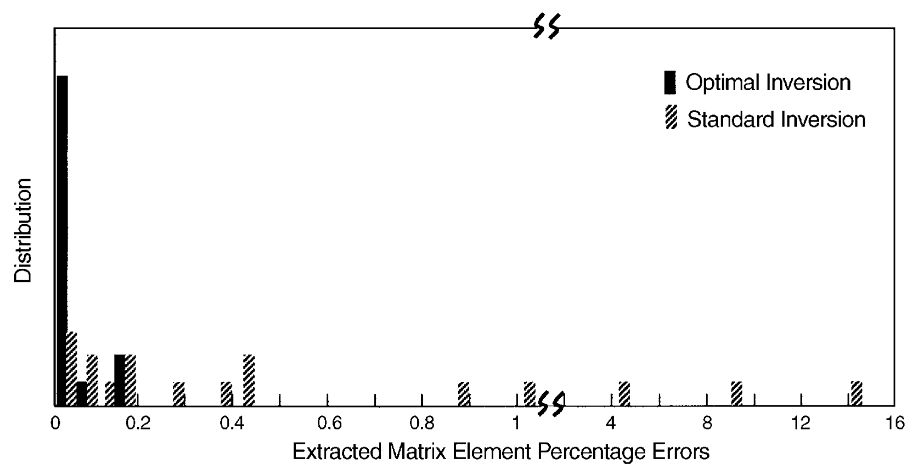

FIG. 2. The distribution of the errors in extracting the dipole matrix elements: A single optimal inversion experiment far outperforms a standard inversion based on 500 random experiments.

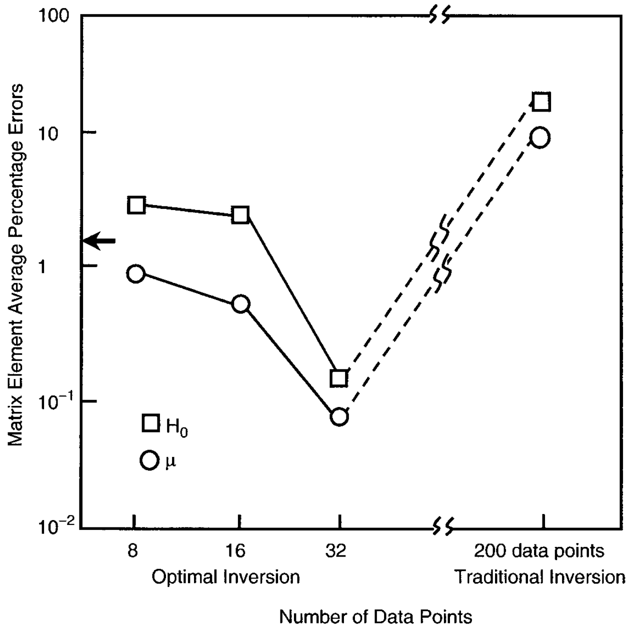

FIG. 3. Quality of the extracted matrix elements for H_0 and μ as a function of the number of simulated recorded data points: The optimal inversion algorithm with only 32 data points is capable of identifying all 64 Hamiltonian matrix elements with errors at least an order of magnitude smaller than the laboratory error of 2% (shown as an arrow on the ordinate). In contrast, a standard nonoptimal inversion with 200 data points produced an unacceptable inversion that magnified the laboratory errors.

phases and amplitudes produced a totally unacceptable inversion, with extracted matrix element errors approximately an order of magnitude larger than the noise in the observations.

Both of these examples indicate that high quality Hamiltonian information may be determined from an optimal set of experiments honed for that specific purpose. The extracted Hamiltonian quality was far better than the noise in the input data. This behavior arises from the closed loop OI machine deducing an optimal control field tailored to the inversion. The data associated with the optimal field combined with the underlying exploratory character of the controlled evolution leaves essentially no freedom for the identified Hamiltonian, except for a narrow distribution around the truth. In both illustrations, operating in a ''standard'' fashion with a very large set of random control experiments produced distinctly inferior inversion results, indicating that the processes involved in achieving OI performance are quite subtle. This point was also evident from examination of some of the fields put aside by the closed loop control algorithm on the way to determining the best final OI experiment; those discarded fields (especially during the early cycles of the loop) and their associated data gave inferior inversion results.

Full synchronization of the loop operations in Fig. 1 is essential, especially to take advantage of the high duty cycle of the laser control experiments [1–4]. In this regard, the most time consuming step in Fig. 1 is likely the data inversion. A variety of techniques may be employed to accelerate this operation, including prior mapping of trial Hamiltonians upon the data [16] and parallelizing the inversion computations. The overall OI concept shows promise as a means for extracting high quality Hamiltonian information, and the concept may be executed with essentially no changes in the present laser control apparatus configurations [17].

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*Present address: California Institute of Technology, Norman Bridge Laboratory of Physics, M.C. 12-33, Pasadena, CA 91125.

†Corresponding author.

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