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***Abstract*—This note considers Hamiltonian identification for a controllable quantum system with non-degenerate transitions and a known initial state. We assume to have at our disposal a single scalar control input and the population measure of only one state at an (arbitrarily large) final time T. We prove that the quantum dipole moment matrix is locally observable in the following sense: for any two close but distinct dipole moment matrices, we construct discriminating controls giving two different measurements. This result suggests that what may appear at first to be very restrictive measurements are actually rich for identification, when combined with well designed discriminating controls, to uniquely identify the complete dipole moment of such systems.**

I. INTRODUCTION

Quantum control has been receiving increasing attention [1] and one of its promising applications is to Hamiltonian identification [2] by using the ability to actively control a quantum system as a means to gain information about the underlying Hamiltonian governing its dynamics. The underlying premise is that controls may be found which make the measurements not only robust to noise but also highly sensitive to the unknown parameters in the Hamiltonian. Hence, although the performance of laboratory measurements may be constrained, the ability to control a quantum system has the prospect of turning this data into a rich source of information on the system's Hamiltonian.

In this note, we consider the problem of identifying the dipole moment (which is assumed to be real) of an $N-$level quantum system, initialized to a known state (ground state), from a single population measurement at one arbitrarily large time T. We suppose an ability to freely control the system with a time dependent electric field $\epsilon(t)$. The measurements are obtained by (i) initializing at time $t = 0$ the system's state at a known state $|i\rangle$, (ii) controlling in open loop and without measurement the system with an electric field $\epsilon_k(t)$ for $t \in [0, T]$ where $T > 0$, and (iii) measuring at final time T the population of one state $|f\rangle$. This may be repeated for many controls $(\epsilon_k)_k$. We prove the existence of controls which make the identification from one population measurement a well posed problem (theorem 1). These controls have a simple physical interpretation in analogy with Ramsey interferometry (see Fig. 1).

The perspective above combined with control theory is motivated by three practical arguments. First, measuring a state population at one time T is a technique which can have a very high signal to noise ratio (~ 100). Second, technological progress with spatial light modulators (SLM) permits generating a broad variety of controls in the laboratory. Third, ultra short pulsed fields can be well measured in the laboratory [3]. Hence, we are able to design a variety of precisely known control inputs.

Le Bris *and al* [4] prove the observability of the dipole moment when the population of all states are measured over an arbitrarily large interval of time. Algorithms to reconstruct the dipole from the measured data were proposed using nonlinear observers [5], [6]. A different setting is considered in [7], [8] where it is supposed that one can prepare and measure the system in a set of orthogonal states at various times, and the available data is the probability to measure the system in a certain state when it was prepared in another; Bayesian estimation is used to reconstruct the energy levels, the damping constants and the dipole moment from the measured data. We consider here the less demanding case where the only available measurement is the population of one state at one arbitrarily large time, and the initial state is known and coincides with the ground state.

The note is organized as follows. In section II we state the main result in Theorem 1, and section III gives the proof of the Theorem and an important lemma on which the main result is based. Finally concluding remarks are presented in Section IV.

II. Observability of the quantum dipole moment

A. Problem setting

We consider a quantum system in a pure state described by the wave function $|\psi\rangle \in \mathcal{S}$. Here $\mathcal{S}$ is the set of N dimensional complex vectors of unit norm. The system interacts with an electric field (the real control input) $\epsilon \in \mathcal{E}_T$ for some $T > 0$ with $\mathcal{E}_T \triangleq \{f : [0,T] \to \mathbb{R} \,|\, f$ piecewise continuous$\}$. For a given control ϵ we measure the population of the state $|f\rangle$ at time T denoted as $P_{if}(\epsilon)$. We denote by H_0 the free Hamiltonian (Hermitian matrix) and by μ the dipole moment operator, also a Hermitian matrix. The initial state $|i\rangle$ and the measured state $|f\rangle$ are eigenvectors of H_0. We consider a semi-classical model for the light-matter interaction, and the dynamics of $|\psi\rangle$ are given by the Schrodinger equation:

$$\begin{aligned} \imath\hbar\frac{\partial}{\partial t}|\psi(t)\rangle &= (H_0 - \epsilon(t)\mu)|\psi(t)\rangle \qquad (1)\\ |\psi(0)\rangle = |i\rangle \quad &, \qquad P_{if}(\epsilon) = |\langle f|\psi(T)\rangle|^2 . \end{aligned}$$

For all $T > 0$, we suppose that we can create any field $\epsilon \in \mathcal{E}_T$ and that we can measure $P_{if}(\epsilon)$. For M different fields $\{\epsilon_1, .., \epsilon_M\}$ we can collect the measurements $\{P_{if}(\epsilon_1), .., P_{if}(\epsilon_M)\}$. Through (1) P_{if} is a function of μ and a functional of ϵ, and when necessary this explicit dependence will be written as $P_{if}(\epsilon, \mu)$. The aim of this note is to explore the feasibility of estimating the dipole moment μ from the measured data $\{P_{if}(\epsilon_1), .., P_{if}(\epsilon_M)\}$ using well chosen controls $\{\epsilon_1, .., \epsilon_M\}$. Below, $P_{if}(\epsilon, \mu)$ refers to the measurement achieved on the real system using a control ϵ, and for any $\hat{\mu}$, $P_{if}(\epsilon, \hat{\mu})$ is the estimated measurement which is obtained by simulating system (1) with the control ϵ and coupling $\hat{\mu}$.

B. Main result

For all $k \leq N$ we denote $|k\rangle$ as the eigenvector of H_0 with associated eigenvalue E_k. Throughout the note, all matrices are written in the basis $(|1\rangle, .., |N\rangle)$. The initial and measured states correspond to some indexes $i, f \in \{1, \ldots, N\}$. For all $k, l \leq N$ we specify $\sigma_{lk}^x \triangleq |l\rangle\langle k| + |k\rangle\langle l|$. We define

$$\mathcal{M} \triangleq \text{Span}\{\sigma_x^{lk} \backslash k, l \leq N \text{ with } \text{Tr}\left(\mu\sigma_x^{lk}\right) \neq 0\},$$

with Tr being the trace operation. When all non diagonal elements of μ are non-null, $M = \dim(\mathcal{M}) = \frac{N(N-1)}{2}$. The main result is the following:

Theorem 1. *Consider a real symmetric matrix μ with zero diagonal entries and a real diagonal matrix H_0 with non-degenerate transitions. Suppose that the system state in (1) is controllable. Then for any positive constant α there exists a time $T > 0$ and M fields $(\epsilon_1, .., \epsilon_M) \in {\mathcal{E}_T}^M$ such that the cost function*

$$J : \mathcal{M} \ni \hat{\mu} \to \sum_{k=1}^{M} \left(P_{if}(\epsilon_k, \hat{\mu}) - P_{if}(\epsilon_k, \mu)\right)^2$$

is in $\mathcal{C}^2(\mathcal{M}, \mathbb{R})$ and locally α-convex[1] *around μ.*

$\mathcal{C}^k(A, B)$ denotes the set of k times continuously differentiable functions defined over A with values in B. In the appendix we provide the definitions of controllability and a matrix with non-degenerate transitions. Here and throughout this note, the norm of matrix μ, noted $\|\mu\|$ refers to the max norm.
A direct consequence of Theorem 1 is the local observability of the dipole moment:

Corollary 1. *Under the assumptions of Theorem 1, the dipole moment is locally observable in $\mathcal{M}$.*

a) Proof: Take $\alpha > 0$. Theorem 1 implies that there exists a time $T > 0$ and M fields $(\epsilon_1, .., \epsilon_M)$ $\in {\mathcal{E}_T}^M$ such that the cost function J is $\mathcal{C}^2(\mathcal{M}, \mathbb{R})$ and locally α-convex around μ. Hence there $\exists r > 0$ such that for all $\hat{\mu} \in \mathcal{M}$ with $\|\hat{\mu} - \mu\| \leq r$ and $\hat{\mu} \neq \mu$, $J(\hat{\mu}) > 0$, and hence there exists $\epsilon \in \{\epsilon_1, .., \epsilon_M\}$ such that $P_{if}(\epsilon, \hat{\mu}) - P_{if}(\epsilon, \mu) \neq 0.\square$

Remark 1. *The local $\alpha-$convexity is a property stronger than the mere possibility to identify the dipole matrix. It states that the distinction between a dipole candidate $\hat{\mu}$ and the true dipole μ can*

[1] The smallest eigenvalue of the Hessian $\nabla^2 J(\mu)$ is larger than α.

be observed (through the measurements aggregated in J) to first order in the distance $\|\mu - \hat{\mu}\|$. This first order dependence of the measurement P_{if} with respect to the dipole μ is addressed in more detail in lemma 1. For well chosen controls, the J function has a very simple shape around μ and a simple gradient algorithm could be used to identify it.

The eigenvalues of H_0 are commonly measured through spectroscopy and can be found in reference tables [9] with precisions of order 10^{-7}. The result of theorem 1 is also relevant for the problem of discriminating between two molecules with the same free Hamiltonian and different effective dipole operators. In that framework, $\hat{\mu}$ and μ would be the dipole operators of these two molecules (as opposed to one estimated and one true dipole, as considered in this note), and the aim is to find controls which produce different data sets for these two different but similar quantum systems. This was experimentally accomplished in [10] where a genetic algorithm is used to find these discriminating controls. A complementary theoretical controllability analysis can be found in [11].

III. Proofs

A. Existence of discriminating controls

We denote $\mu' = \frac{1}{\|\mu\|}\mu$ the normalized dimensionless dipole moment operator, $\mu'_{lk} \triangleq \operatorname{Tr}\left(\mu' \sigma_x^{lk}\right)$ and $\frac{\partial P_{if}}{\partial \mu'_{lk}}(\epsilon)$ the partial derivative of $P_{if}(\epsilon)$ with respect to μ'_{lk}. Theorem 1 is based on the following lemma:

Lemma 1. *Suppose that μ is real, symmetric and has only zeros on its diagonal and H_0 is real, diagonal, with non-degenerate transitions. Suppose system* (1) *is controllable. Then for all (l,k) with $\mu_{lk} \neq 0$, there exists $\xi_0 > 0$ such that, for all $\xi \in]0, \xi_0[$, exist $T > 0$ and $\epsilon \in \mathcal{E}_T$ satisfying*

- $\frac{\partial P_{if}}{\partial \mu'_{lk}}(\epsilon) = \frac{1}{2\xi} + O(1)$
- $\forall \{m,n\} \neq \{l,k\}$ *with* $\mu_{mn} \neq 0$, $\frac{\partial P_{if}}{\partial \mu'_{mn}}(\epsilon) = O(1)$,

where $O(1)$ corresponds to zero order terms with respect to ξ around 0^+.

B. Proof of Theorem 1

To each pair of integers (l_p, k_p), $l_p < k_p$ such that $\operatorname{Tr}\left(\mu' \sigma_x^{l_p k_p}\right) \neq 0$ we associate a unique index $p \in \{1, ..., M\}$, and we define $\sigma_x^p \triangleq \sigma_x^{l_p k_p}$ along with $\mu'_p \triangleq \operatorname{Tr}(\mu' \sigma_x^p)$.

According to lemma 1, $\exists \xi_0 > 0$ such that $\forall \xi \in]0, \xi_0[$, $\exists\, T_1, .., T_M$ and $(\epsilon_1, .., \epsilon_M) \in \mathcal{E}_{T_1} \times .. \times \mathcal{E}_{T_M}$ such that: (i) $\forall p \in [1:M]$ $\frac{\partial P_{if}}{\partial \mu'_p}(\epsilon_p) = \frac{1}{2\xi} + O(1)$ and (ii) $\forall p' \neq p$ $\frac{\partial P_{if}}{\partial \mu'_{p'}}(\epsilon_p) = O(1)$. We take $T = \max(T_1, .., T_M)$ and for all $k \in \{1, .., M\}$ we extend the definition of ϵ_k from $[0, T_k]$ to $[0, T]$ by taking $\epsilon_k(t) = 0$ for all $t \in]T_k, T]$. We will use $J : \mathcal{M} \to \mathbb{R}$ defined by:

$$J(\hat{\mu}) = \sum_{k=1}^{M} \left(P_{if}(\epsilon_k, \hat{\mu}) - P_{if}(\epsilon_k, \mu)\right)^2.$$

For a fixed $T > 0$ and $\epsilon \in \mathcal{E}_T$, since $\mu \to P_{if}(\epsilon, \mu)$ is $\mathcal{C}^1(\mathcal{M}, \mathbb{R})$, $J(\hat{\mu})$ is in $\mathcal{C}^2(\mathcal{M}, \mathbb{R})$. We find $\frac{\partial^2 J}{\partial \mu'_p \partial \mu'_{p'}}(\mu) = \sum_{k=1}^{M} \frac{\partial P_{if}}{\partial \mu'_p}(\epsilon_k, \mu) \frac{\partial P_{if}}{\partial \mu'_{p'}}(\epsilon_k, \mu)$ so that for all $p \in \{1, ..., M\}$: $\frac{\partial^2 J}{\partial {\mu'_p}^2}(\mu) = \frac{1}{4\xi^2} + O(\frac{1}{\xi})$ and when $p \neq p'$ $\frac{\partial^2 J}{\partial \mu'_p \partial \mu'_{p'}}(\mu) = O(\frac{1}{\xi})$. We have: $\nabla^2 J(\mu) = \frac{1}{4\xi^2}\left(I + O(\xi)\right)$, where $\nabla^2 J(\mu)$ is the Hessian of J at μ and I is the identity matrix. The smallest eigenvalue of $\nabla^2 J(\mu)$ scales as $\frac{1}{4\xi^2}(1 + O(\xi))$, hence by taking ξ small enough it can be made larger than α thereby reaching the conclusion above. $\square$

C. Proof of lemma 1

We define the dimensionless time scale $\tau \triangleq \frac{1}{\hbar}\|H_0\| t$ and also $\top \triangleq \frac{1}{\hbar}\|H_0\| T$. For two times $\tau, \tau' \in [0, \top]$ we define the propagator $U(\tau', \tau)$ such that $|\psi(\tau')\rangle = U(\tau', \tau)|\psi(\tau)\rangle$. Rewriting (1) for $U(\tau, 0)$ we obtain:

$$\begin{aligned} \imath \frac{\partial}{\partial \tau} U(\tau, 0) &= \frac{1}{\|H_0\|}(H_0 - \epsilon(\tau)\mu) U(\tau, 0) \qquad (2) \\ P_{if}(\epsilon) &= |\langle f| U(\top, 0) |i\rangle|^2, U(0,0) = I\ . \end{aligned}$$

The proof of lemma 1 has two parts I and II separately treated below.

i) Part I: Take two times τ_1, τ_2, $0 < \tau_1 < \tau_2 < \top$. We can write (for any complex z we denote by $\bar{z}$ its complex conjugate): $P_{if}(\epsilon) = z\bar{z}$ where $z = \langle f| U(\top, \tau_2) U(\tau_2, \tau_1) U(\tau_1, 0) |i\rangle$.

Denote for any $m, n = 1, ..., M$: $\omega'_{mn} \triangleq \frac{E_m - E_n}{\|H_0\|}$ and consider the control defined on $[\tau_1, \tau_2]$:

$$\epsilon(\tau) = \varepsilon \cos(\omega'_{lk}(\tau - \tau_1))\ , \qquad (3)$$

where ε is a small strictly positive real parameter. Take $\xi = \frac{\|\mu\|\varepsilon}{\|H_0\|}$. The only remaining degree of

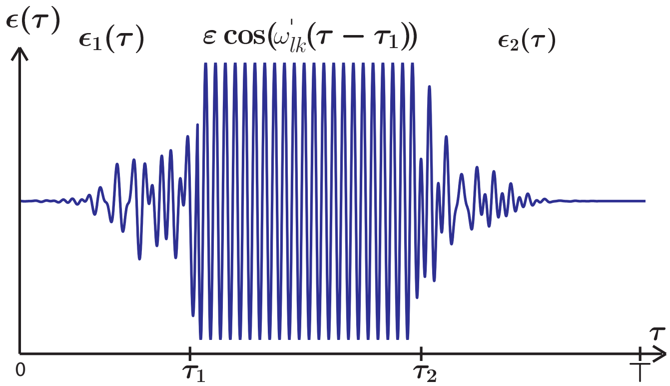

Fig. 1. A *good* control ϵ has three components (inspired from Ramsey interferometry) to enable the identification of μ_{lk}. The field ϵ_1 is defined over $[0,\tau_1]$ (analog of first Ramsey pulse) to steer the known initial state $|i\rangle$ to $|\psi_1\rangle = |l\rangle$: $|l\rangle = U(\tau_1, 0)\,|i\rangle$. The field ϵ_2 is defined over $[\tau_2, \top]$ (analog of second Ramsey pulse) is such that $|f\rangle = U(\top, \tau_2)\,|\psi_2\rangle$ where $|\psi_2\rangle = U(\tau_2,\tau_1)\left(\frac{|l\rangle + \imath|k\rangle}{\sqrt{2}}\right)$ and the propagator $U(\tau_2,\tau_1)$ corresponds, for a long interval $\tau_2-\tau_1$, to a large number of Rabi oscillations with the control $\varepsilon\cos(\omega'_{lk}(\tau-\tau_1))$ resonant with the $|l\rangle \leftrightarrow |k\rangle$ transition.

freedom in the control over $[\tau_1,\tau_2]$ is ξ, which can be taken arbitrarily small. We define $H'_0 = \frac{1}{\|H_0\|}H_0$ and $\omega'_{mn} = \langle m|\,H'_0\,|m\rangle - \langle n|\,H'_0\,|n\rangle$. Note that $\omega'_{mn} = -\omega'_{nm}$. We have [12]

$$\frac{\partial}{\partial\mu'_{lk}}U(\tau_2,\tau_1) = \imath\frac{\|\mu\|}{\|H_0\|}U(\tau_2,\tau_1) \times \int_{\tau_1}^{\tau_2}\epsilon(\tau)U^\dagger(\tau,\tau_1)\sigma_x^{lk}U(\tau,\tau_1)d\tau\ . \quad (4)$$

We now rewrite (2) and (4) for the control given in (3) on the time interval $[\tau_1,\tau_2]$:

$$\imath\frac{\partial}{\partial\tau}U(\tau,\tau_1) = (H'_0 - \xi\cos(\omega'_{lk}(\tau-\tau_1))\mu')U(\tau,\tau_1) \quad (5)$$

$$\frac{\partial}{\partial\mu'_{lk}}U(\tau_2,\tau_1) = \imath\xi U(\tau_2,\tau_1) \times \int_{\tau_1}^{\tau_2}\cos(\omega'_{lk}(\tau-\tau_1))U^\dagger(\tau,\tau_1)\sigma_x^{lk}U(\tau,\tau_1)d\tau\ . \quad (6)$$

The goal is to show that $\frac{\partial}{\partial\mu'_{lk}}U(\tau_2,\tau_1)$ can be made arbitrarily "large" while $\frac{\partial}{\partial\mu'_{mn}}U(\tau_2,\tau_1)$ stays bounded. Note that all the terms in the integrand of (6) are bounded, and a rough estimate of the norm of $\frac{\partial}{\partial\mu'_{lk}}U(\tau_2,\tau_1)$ gives a quantity proportional to $(\tau_2-\tau_1)\xi$. Hence, we take $\tau_2-\tau_1 = \frac{1}{\xi^2}$, implying the need to have expressions for $U(\tau,\tau_1)$ over a time scale on the order of $\frac{1}{\xi^2}$. To this end we state lemma 2 which gives such an approximation.

Lemma 2. *Consider Eq.* (5). *There exists a Hermitian matrix K and $\xi_0 > 0$ such that, for any $\xi \in]0,\xi_0[$, we have:*

$$\sup_{\tau\in[\tau_1,\tau_1+\frac{1}{\xi^2}]}\|U(\tau,\tau_1) - e^{-\imath H'_0(\tau-\tau_1)}e^{\imath(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\xi^2K)(\tau-\tau_1)}\| = O(\xi).$$

We continue with the proof of Lemma 1 and will come back to Lemma 2 in Section III-D.

Using the expression of $U(\tau,\tau_1)$ given in lemma 2, the integrand in (6) is:

$$\begin{aligned}&\cos(\omega'_{lk}(\tau-\tau_1))U^\dagger(\tau,\tau_1)\sigma_x^{lk}U(\tau,\tau_1) = \\ &\quad e^{-\imath(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\xi^2K)(\tau-\tau_1)}(\cos(\omega'_{lk}(\tau-\tau_1))e^{\imath H'_0(\tau-\tau_1)} \\ &\qquad \sigma_x^{lk}e^{-\imath H'_0(\tau-\tau_1)})e^{\imath(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\xi^2K)(\tau-\tau_1)} + O(\xi)\ .\end{aligned}$$

In order to compute (6), we need the following result:

$$\begin{aligned}&\cos(\omega'_{lk}(\tau-\tau_1))e^{\imath H'_0(\tau-\tau_1)}\sigma_x^{lk}e^{-\imath H'_0(\tau-\tau_1)} = \\ &\quad \tfrac{1}{2}\sigma_x^{lk} + \tfrac{1}{2}\cos(2\omega'_{lk}(\tau-\tau_1))\sigma_x^{lk} \\ &\qquad + \tfrac{1}{2}\sin(2\omega'_{lk}(\tau-\tau_1))\sigma_y^{lk}\ , \quad (7)\end{aligned}$$

where we denote $\sigma_y^{lk} = +\imath\,|l\rangle\langle k| - \imath\,|k\rangle\langle l|$. In (7), the terms oscillating at frequency $2\omega'_{lk}$ independent of ξ will only contribute to $O(\xi)$ in (6). We now focus on the contribution of the term with σ_x^{lk} in (6) which calls for (see appendix): $\forall\tau$

$$e^{-\imath(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\xi^2K)(\tau-\tau_1)}\sigma_x^{lk}e^{\imath(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\xi^2K)(\tau-\tau_1)} = \sigma_x^{lk} + O(\xi)\ . \quad (8)$$

Introducing (8) into (6), we find:

$$\frac{\partial}{\partial\mu'_{lk}}U(\tau_2,\tau_1) = \imath\xi U(\tau_2,\tau_1) \left(\frac{\tau_2-\tau_1}{2}\sigma_x^{lk} + O(1) + (\tau_2-\tau_1)O(\xi)\right)\ .$$

From now on, we take $\tau_2 = \tau_1 + \frac{1}{\xi^2}$ and obtain:

$$\frac{\partial}{\partial\mu'_{lk}}U(\tau_2,\tau_1) = \imath U(\tau_2,\tau_1)(\tfrac{1}{2\xi}\sigma_x^{lk} + O(1)). \quad (9)$$

We define $|\psi_1\rangle \triangleq |l\rangle$ and $|\psi_2\rangle \triangleq \frac{1}{\sqrt{2}}U(\tau_2,\tau_1)(|l\rangle + \imath\,|k\rangle)$. Since the system is controllable there exists a time τ_1 and a field $\epsilon_1 \in \mathcal{E}_{\tau_1}$ such that $U(\tau_1,0)\,|i\rangle = |\psi_1\rangle$, and there exists a time $\top$ and a field ϵ_2 defined over $[\tau_2,\top]$ such that $U^\dagger(\top,\tau_2)\,|f\rangle = |\psi_2\rangle$. Since the state space is compact (here it is a sphere), we know that if the system is controllable, it is controllable in bounded time, and with bounded controls (see Theorem 6.5 in [13]). Hence, $\top - \tau_2$ can be chosen bounded for all ξ. Therefore $\frac{\partial}{\partial\mu_{lk}}U(0,\tau_1)$ and $\frac{\partial}{\partial\mu_{lk}}U(\tau_2,\top)$ are bounded. Thus, we have:

$$\frac{\partial}{\partial\mu'_{lk}}P_{if}(\epsilon)=2\Re(\langle f|\,U(\top,\tau_2)\frac{\partial}{\partial\mu'_{lk}}U(\tau_2,\tau_1)U(\tau_1,0)\,|i\rangle \\ \langle i|\,U^\dagger(\tau_1,0)U^\dagger(\tau_2,\tau_1)U^\dagger(\top,\tau_2)\,|f\rangle)+O(1).$$

We now utilize $U(\tau_1,0)\,|i\rangle = |\psi_1\rangle$ and $U^\dagger(\top,\tau_2)\,|f\rangle = |\psi_2\rangle$ where $|\psi_1\rangle$ and $|\psi_2\rangle$ are defined above, and replace $\frac{\partial}{\partial\mu'_{lk}}U(\tau_2,\tau_1)$ by its expression in (9) to find: $\frac{\partial}{\partial\mu'_{lk}}P_{if}(\epsilon)=\frac{1}{2\xi}+O(1)$. This expression holds for the control defined as:

$$\epsilon(\tau)=\begin{cases}\epsilon_1(\tau), & \text{if } \tau\in[0,\tau_1]\\ \frac{\|H_0\|}{\|\mu\|}\xi\cos(\omega'_{lk}(\tau-\tau_1)), & \text{if } \tau\in]\tau_1,\tau_2[\\ \epsilon_2(\tau), & \text{if } \tau\in[\tau_2,\top]\end{cases} \tag{10}$$

ii) Part II: We now need to prove that $\frac{\partial}{\partial\mu'_{mn}}P_{if}(\epsilon)=O(1)$ for $\{m,n\}\neq\{l,k\}$, where ϵ is the control found above in (10). As in Eq. (6), we have:

$$\frac{\partial}{\partial\mu'_{mn}}U(\tau_2,\tau_1)=\imath\xi U(\tau_2,\tau_1)\times \\ \int_{\tau_1}^{\tau_2}\cos(\omega'_{lk}(\tau-\tau_1))U^\dagger(\tau,\tau_1)\sigma_x^{mn}U(\tau,\tau_1)d\tau\ , \tag{11}$$

and again the result of lemma 2 is employed. Eq. (11) calls for

$$2\cos(\omega'_{lk}(\tau-\tau_1))e^{\imath H'_0(\tau-\tau_1)}\sigma_x^{mn}e^{-\imath H'_0(\tau-\tau_1)}= \\ \cos((\omega'_{lk}-\omega'_{mn})(\tau-\tau_1))\sigma_x^{mn}-\sin((\omega'_{lk}-\omega'_{mn})(\tau-\tau_1))\sigma_y^{mn}+ \\ \cos((\omega'_{lk}+\omega'_{mn})(\tau-\tau_1))\sigma_x^{mn}+\sin((\omega'_{lk}+\omega'_{mn})(\tau-\tau_1))\sigma_y^{mn}. \tag{12}$$

Considering that H_0 has non-degenerate transitions (see definition in the appendix) implies that $\omega'_{lk}-\omega'_{mn}\neq 0$ and $\omega'_{lk}+\omega'_{mn}\neq 0$. As the expression in (12) oscillates at frequencies independent of ξ, it therefore contributes to $O(\xi)$ in (11). Hence, for $\tau_2-\tau_1=\frac{1}{\xi^2}$ we can directly conclude that $\frac{\partial}{\partial\mu'_{mn}}P_{if}(\epsilon)=O(1)$. □

D. Proof of lemma 2

This proof relies on three consecutive changes of frame that aim to cancel the oscillating terms of order 0 and 1 with respect to ξ. We then derive a specific form of the averaging Theorem (see theorem 4.3.6 in [14] for a general form of the averaging theorem). For the sake of clarity and with no loss of generality, we take $\tau_1=0$ and note $U(\tau)\triangleq U(\tau,\tau_1)$. Eq. (5) may be written in the interaction frame $U_I(\tau)\triangleq e^{\imath H'_0\tau}U(\tau)$,

$$\frac{\partial}{\partial\tau}U_I(\tau) = \imath\xi\left(\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\frac{\partial}{\partial\tau}H_I(\tau)\right)U_I(\tau)$$

where:

$$\frac{\partial}{\partial\tau}H_I(\tau)=\tfrac12\sum_{(m,n)\neq(k,l)}\mu'_{mn}e^{\imath(-\omega'_{kl}+\omega'_{mn})\tau}\,|m\rangle\langle n| \\ +\tfrac12\sum_{(m,n)\neq(l,k)}\mu'_{mn}e^{\imath(-\omega'_{lk}+\omega'_{mn})\tau}\,|m\rangle\langle n|\ ,$$

and the average of H_I is zero. The average of a time dependent operator $C(\tau)$ is defined as follows (see defintion 4.2.4 in [14]): $\overline{C}=\lim_{\theta\to+\infty}\frac{1}{\theta}\int_0^\theta C(\tau)d\tau$. We now take $U'_I(\tau)=(I-\imath\xi H_I(\tau))U_I(\tau)$. Since $\frac{\partial}{\partial\tau}H_I$ is almost periodic[2], then H_I is also almost periodic and hence bounded for all τ. Hence, there exists $\xi_0>0$, $\forall\xi<\xi_0$, $I-\imath\xi H_I(\tau)$ has an inverse and $(I-\imath\xi H_I(\tau))^{-1}=I+\imath\xi H_I(\tau)+O(\xi^2)$. We find:

$$\frac{\partial}{\partial\tau}U'_I(\tau)=\imath\left(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}- \right. \\ \left.\imath\xi^2\left(\frac{\mu'_{lk}}{2}[H_I(\tau),\sigma_x^{lk}]+H_I(\tau)\frac{\partial}{\partial\tau}H_I(\tau)\right)+O(\xi^3)\right)U'_I(\tau).$$

Notice that, with $K=-\imath\overline{H_I\frac{\partial}{\partial\tau}H_I}$ independent of ξ and $\tilde{K}(\tau)$ almost periodic with zero average, we also have: $\frac{\mu'_{lk}}{2}[H_I(\tau),\sigma_x^{lk}]+H_I(\tau)\frac{\partial}{\partial\tau}H_I(\tau)=\imath(K+\frac{\partial}{\partial\tau}\tilde{K}(\tau))$. It is important to note that $\frac12\overline{\frac{\partial}{\partial\tau}H_I^2}=0=\overline{H_I\frac{\partial}{\partial\tau}H_I+(\frac{\partial}{\partial\tau}H_I)H_I}=\imath(K-K^\dagger)$. Hence $K=K^\dagger$ is Hermitian.

We now take $U''_I(\tau)=(I-\imath\xi^2\tilde{K}(\tau))U'_I(\tau)$. Since $\tilde{K}(\tau)$ is bounded for all τ, then for a sufficiently small ξ, $I-\imath\xi^2\tilde{K}(\tau)$ has an inverse and $(I-\imath\xi^2\tilde{K}(\tau))^{-1}=I+\imath\xi^2\tilde{K}(\tau)+O(\xi^4)$. U''_I satisfies the following equation:

$$\frac{\partial}{\partial\tau}U''_I(\tau)=\imath\left(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\xi^2K+O(\xi^3)\right)U''_I(\tau)\ , \tag{13}$$

and we define U_{av} to be the solution to the averaged dynamics ($U_{av}(0)=I$):

$$\frac{\partial}{\partial\tau}U_{av}(\tau)=\imath\left(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\xi^2K\right)U_{av}(\tau)\ . \tag{14}$$

We can directly solve (14): $U_{av}(\tau)=e^{\imath\left(\xi\frac{\mu'_{lk}}{2}\sigma_x^{lk}+\xi^2K\right)\tau}$. Subtracting (13) from (14), we find, using Gronwall's lemma, that for all $\tau<\frac{1}{\xi^2}$ one has $U''_I(\tau)=U_{av}(\tau)+O(\xi)$. Also note that to go from U_I to U''_I we have used two

[2]Can be written as $\sum_{k=1}^M e^{i\omega_k\tau}A_k$

consecutive changes of variables which are close to the identity, hence: $\forall\tau\ U_I''(\tau) = U_I(\tau) + O(\xi)$. Finally, since $e^{-\imath H_0'\tau}$ is an isometry, we have:

$$U(\tau) = e^{-\imath H_0'\tau} e^{\imath(\xi \frac{\mu_{lk}'}{2}\sigma_x^{lk} + \xi^2 K)\tau} + O(\xi) \text{ for all } \tau \leq \frac{1}{\xi^2}.\square$$

IV. Conclusion

Identification of the real dipole moment matrix is shown to be well posed for a controllable finite dimensional quantum system with non-degenerate transitions and using as measurements only one population at a final time T. The results also provide a theoretical foundation to optimal discrimination experiments.

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Appendix

Definition 1. *We say that system* (1) *is controllable [15] if for all* $|\psi_1\rangle, |\psi_2\rangle \in \mathcal{S}$ *there exists a time* t *and a control* $\epsilon \in \mathcal{E}_t$ *such that for* $|\psi(0)\rangle = |\psi_1\rangle$, (1) *leads to* $|\psi(t)\rangle = |\psi_2\rangle$.

Definition 2. *Let* H_0 *and* μ *be* $N \times N$ *Hermitian matrices. We denote* $E_1, .., E_N$ *the eigenvalues of* H_0 *and* $|1\rangle, .., |N\rangle$ *its corresponding eigenvectors. We say that* H_0 *has non-degenerate transitions [16] if* $\forall (l,k) \neq (m,n)$, $l \neq k$ *and* $m \neq n$, *such that* $\langle l|\mu|k\rangle \neq 0$ *and* $\langle m|\mu|n\rangle \neq 0$, *we have* $E_l - E_k \neq E_m - E_n$.

Definition 3. *Take system* (1). *Let us denote* $\mathcal{M}$ *as the space to which* μ *belongs. We say that* μ *is locally observable in* $\mathcal{M}$ *if there exists* $r > 0$ *such that for all* $\hat{\mu} \in \mathcal{M}$ *with* $0 < \|\hat{\mu} - \mu\| \leq r$ *there exists* $T > 0$ *and* $\epsilon \in \mathcal{E}_T$ *such that* $P_{if}(\epsilon, \hat{\mu}) \neq P_{if}(\epsilon, \mu)$.

Computation: Here, we compute $\Sigma_x^{lk}(\tau) = e^{-i(\xi\frac{\mu_{lk}'}{2}\sigma_x^{lk} + \xi^2 K)(\tau-\tau_1)} \sigma_x^{lk} e^{i(\xi\frac{\mu_{lk}'}{2}\sigma_x^{lk} + \xi^2 K)(\tau-\tau_1)}$. We have $\mu_{lk}' \neq 0$ and $\sigma_x^{lk} + \xi\frac{2K}{\mu_{lk}'}$ is Hermitian. Hence, there exists a unitary matrix P_ξ and a real diagonal matrix Δ_ξ such that $\sigma_x^{lk} + \xi\frac{2K}{\mu_{lk}'} = P_\xi \Delta_\xi P_\xi^\dagger$. The function $\xi \in [0, \xi_0] \to \sigma_x^{lk} + \xi\frac{2K}{\mu_{lk}'}$ is analytic, therefore the eigenvectors of $\sigma_x^{lk} + \xi\frac{2K}{\mu_{lk}'}$ can be continued analytically as a function of ξ (see Theorem 6.1 in chapter II, §6 section 1 and 2 in [17]). Hence, $P_\xi = P_0 + O(\xi)$ where P_0 is such that $P_0^\dagger \sigma_x^{lk} P_0 = \sigma_z^{lk}$ is real and diagonal. $\sigma_z^{lk} = |l\rangle\langle l| - |k\rangle\langle k|$. We find, $\forall\tau$: $\Sigma_x^{lk}(\tau) = \sigma_x^{lk} + O(\xi)$, where $O(\xi)$ is a first order term in ξ and a bounded function of τ.