Observer-based Hamiltonian identification for quantum systems ⋆

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Abstract

A symmetry-preserving observer-based parameter identification algorithm for quantum systems is proposed. Starting with a qubit (2-level quantum system) and where the unknown parameters consist of the atom-laser frequency detuning and coupling constant, we prove an exponential convergence result. The analysis is inspired by the Lyapounov and adaptive control techniques and is mainly based on the averaging arguments and some relevant transformations. The observer is then extended to the multi-level case where eventually all the atom-laser coupling constants are unknown. The extension of the convergence analysis is discussed through some heuristic arguments. The relevance and the robustness with respect to various noises are tested through numerical simulations.

Key words: Quantum systems, Nonlinear systems, Asymptotic observers, Symmetries, Averaging.

1 Introduction

The ability of coherent light to manipulate molecular systems at the quantum scale has been demonstrated both theoretically and experimentally [2, 22, 39, 3, 4, 33, 35, 22, 7, 40, 25, 23, 20, 32, 28, 27]. Many of the procedures, considered in this aim, are based on the possibility to perform a large number of experiments in a very small time frame. Thus, the output provided by these experiments can be used to correct the process and to identify more satisfactory control fields [14, 32, 23].

The ability to rapidly generate a large amount of quantum dynamics data may also be used to extract more information about the possibly unknown parameters of the quantum system itself. For each test field (i.e., control), there is the possibility of performing many observations for deducing information about the system, and this process can often be carried out at a much faster rate than the associated numerical simulations of the dynamics. Moreover, the recent advances in laser technology provide the means for generating a very large class of test fields for such experiments.

The rapidly developing theory of quantum parameter estimation has been investigated through different approaches. The maximum-likelihood methods and the subsequent experiment design techniques provide a first class of results in this area [24, 31, 16, 17, 18]. maximum-entropy methods [6] and maximum Kullback entropy methods [29] provide a second class of parameter estimation algorithms for quantum systems. The optimal identification techniques via least-square criteria's [10, 9, 21] and the map inversion techniques [36] are some other techniques explored in this area. Finally Kalman filtering techniques [11, 37] have been applied to some atomic magnetometery problems.

However, in general, developing effective identification algorithms is of a great interest in this domain. The main concerns in quantum parameter estimation theory and in particular the above methods are the presence of local minima for the optimization problems, sensitivity with respect to the experimental uncertainties and noises and finally the heavy cost of computations in formerly developed algorithms.

Before going through the identification and the experiment design problems, we need to ensure the identifiability of the system. This issue has been addressed in a recent work [21] where sufficient assumptions applying the uniqueness of the inversion result are provided in two relevant settings. A brief review of an identifiability result needed for the purpose of this paper is given in the Appendix B. The semi-constructive proof in [21] suggests that a well-chosen control laser field, coupling all the eigenstates of the free Hamiltonian, would be sufficient to identify the unknown parameters.

In [15], a state observer for a known quantum system is pro-

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posed. This observer is then used as a basis for the quantum parameter estimation applying an iterative search algorithm. The provided optimization algorithms typically converge toward local minima.

Here, in the same direction, we provide an observer-based parameter estimation algorithm based on techniques derived from adaptive control theory. In this aim, we will integrate online a generalized observer including the estimators for both the unknown state and the unknown parameters of the system. The main interest of this asymptotic observer with respect to the previous methods relies on its non-linearity preserving the system structure, on its small cost of computations and robustness with respect to uncertainties, as shown by the simulations. The local convergence result (theorem 1) is based on the local and exponential convergence of the averaged system (26) which has a large basin of attraction due to its triangular structure. Moreover, the convergence of the estimation of the parameter $\hat{\mu}$ (coupling constant) is global for the averaged system. Such global convergence properties (for $\hat{\mu}$) ensure in practice a large domain of convergence for the non-linear asymptotic observer as shown by simulations.

We start as a simple quantum system, with a two level system described by a Schrödinger dynamics and a time-continuous population measurement. We propose an algorithm based on nonlinear asymptotic observer techniques preserving the symmetries ([5]) to estimate the system parameters. As far as we know, such recent techniques have not been applied to tackle this problem and this paper illustrates their potential interest. Since the observer design exploits the physical symmetries (invariance with respect to the frame-change), the filter equation admits a natural physical and geometrical interpretation that can be extended to higher dimensions. This extension will be addressed in Section 5.

In Section 2, we provide an observer which estimates the qubit wave function (in fact we rather use the density matrix language) and the two parameters Δ and μ at the same time. The efficiency and the robustness of the estimator are tested through some simulations in the same section. In Section 3, the structure and the design technique of the proposed observer are analyzed. The Section 4 is dedicated to the proof of a convergence result for the estimation algorithm. In Section 5, we propose an extension of the observer to the multi-level systems where an exact knowledge of the frequencies (no detuning) and the only parameters to identify are the atom-laser coupling constants. After testing the relevance and the robustness of the observer on a 3-level system, we discuss a heuristic extension of the convergence proof to the multi-level case.

We use the standard notations in quantum physics literature (see e.g. [8], and [13] for a more advanced lesson).

2 Observer-based Qubit Hamiltonian identification

2.1 Dynamics

The Schrödinger equation for the system writes:

$$\imath\frac{d}{dt}\Psi = \left(\frac{\Delta}{2}\sigma_z + \frac{u\mu}{2}\sigma_x\right)\Psi, \qquad \Psi = \begin{pmatrix}\Psi_1\\ \Psi_2\end{pmatrix} \in \mathbb{C}^2 \qquad (1)$$

where

$$\sigma_x = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix}, \quad \sigma_y = \begin{pmatrix}0 & -i\\ i & 0\end{pmatrix}, \quad \sigma_z = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}$$

denote the Pauli matrices, Δ is the difference between the atomic transition frequency (of ground state to excited state $\omega_{e0} = \omega_{a,e} - \omega_{a,0}$) and the laser frequency ω, μ is the atom-laser coupling constant and $u(t) \in \mathbb{R}$ is the slowly varying amplitude of the laser. We have $\sigma_x^2 = 1$; $\sigma_x\sigma_y = \imath\sigma_z$ (the preceding identity holds for any circular permutation of the indices x, y, and z), the output is

$$y = <\sigma_z\Psi, \Psi> = |\Psi_1|^2 - |\Psi_2|^2 = 2|\Psi_1|^2 - 1$$

as the measurement is the ground state population $|\Psi_1|^2$ and the conservation of probability implies $|\Psi_1|^2 + |\Psi_2|^2 = 1$.

Here, let us give an idea of the physical setup leading to the above 2-level model. Note that a similar multi-level counterpart can be considered, as well. The physical setup consists of an ensemble of identically prepared systems undergoing the same dynamics: for instance, dilute mono-atomic gases. They are very simple systems, in the sense their constituents (atoms), are perfectly identical and interact very weakly with each other. Atoms in such gases can be considered as perfect quantum systems, with a sequence of discrete energy states labeled $|i\rangle$, for $i \in \mathbb{N}$, with increasing energies $E_i = \hbar\omega_{a,i}$ depending only on the atomic species considered. In order to measure the population of the ground state (i.e, the state of the lower energy), the system is illuminated with coherent light (a first laser) whose frequency is close to the transition frequency corresponding to the energy gap $E_j - E_0 = \hbar(\omega_{a,j} - \omega_{a,0})$ of a very unstable excited state (i.e, having a very short lifetime) to the ground state transition. It generates a transition $|0\rangle \to |j\rangle$ for a part of the population illuminated from the ground state to the excited state, which spontaneously decays to the ground state emitting a photon. The measurement of the number of photons emitted is then directly proportional to the population of the ground state $|0\rangle$. Suppose there is also another laser whose frequency is close to the transition frequency of another excited state $|e\rangle$ to the ground state $|0\rangle$. The lifetime of the transition between the two latter states is supposed to be much longer than the previous one. To a first approximation the dynamics of the two-state system (ground state $|0\rangle$ and excited state $|e\rangle$ having the longest lifetime) is described by a Schrödinger equation. We assume that some parameters are not well known: Δ which is the difference between the second laser frequency and the atomic transition frequency, μ which is the atom-laser coupling strength and characterizes the Rabi frequency. The goal is to identify in real time Δ and μ, measuring the ground state population thanks to the first laser (which generates a transition only for ground state population) and the photo-detector. The usual modeling of these open-quantum systems via Lindbald type terms in the density matrix dynamics (see, e.g., [13]) is analyzed in [26], where singular perturbation techniques are applied to justify the adiabatic (quasi-static) approximations usually made by physicists and leading to a dynamical model described (up to higher order terms) by the Schrödinger equation (1) despite a continuous population measurement. Moreover, note that the well-posedness of the identification problem for the above system may be deduced from the Appendix B.

It is convenient to write the dynamics with the density matrix: let $\rho = \Psi\Psi^\dagger$ denote the complex matrix associated to the projector on the state Ψ. Supposing that the system is pure (meaning it is not entangled to its environment) implies both properties $Tr(\rho) = \Psi_1^\dagger\Psi_1 + \Psi_2^\dagger\Psi_2 = 1$ and $\rho^2 = \Psi\Psi^\dagger\Psi\Psi^\dagger = \rho$. Thus rewriting (1) the system becomes

$$\frac{d}{dt}\rho = -\imath\left[\frac{\Delta}{2}\sigma_z + \frac{u\mu}{2}\sigma_x, \rho\right] \tag{2}$$

$$\frac{d}{dt}\mu = 0 \tag{3}$$

$$\frac{d}{dt}\Delta = 0 \tag{4}$$

$$y = Tr(\sigma_z\rho) \tag{5}$$

where [,] is the commutator. We assume the laser amplitude to be slowly varying compared to the Rabi frequency $|u\mu|$, i.e., $|\dot{u}| << |u\mu||u|$ (the Rabi frequency is a characteristic of the absorption-emission cycle of photons for an illuminated atom). We assume, moreover, that the frequencies of the laser and the frequencies of the atomic transition are close: $|\Delta| << |u|\mu$.

2.2 *The adaptive observer*

Consider the adaptive observer

$$\frac{d}{dt}\hat{\rho} = -\imath\left[\frac{\hat{\Delta}}{2}\sigma_z + \frac{u\hat{\mu}}{2}\sigma_x, \hat{\rho}\right] - K_\rho\left(\mathrm{Tr}(\sigma_z\hat{\rho}) - y\right)\left(\sigma_z\hat{\rho} + \hat{\rho}\sigma_z - 2\mathrm{Tr}(\sigma_z\hat{\rho})\hat{\rho}\right) \tag{6}$$

$$\frac{d}{dt}\hat{\mu} = -uK_\mu\mathrm{Tr}(\sigma_y\hat{\rho})\left(\mathrm{Tr}(\sigma_z\hat{\rho}) - y\right) \tag{7}$$

$$\frac{d}{dt}\hat{\Delta} = -uK_\Delta\mathrm{Tr}(\sigma_x\hat{\rho})\left(\mathrm{Tr}(\sigma_z\hat{\rho}) - y\right) \tag{8}$$

where K_ρ, K_μ and K_Δ are positive scalars. As we did for the true system, let us suppose that u is constant, and $|\hat{\Delta}| \ll |u\mu|$. Suppose the order of magnitude of μ is known initially: $|\hat{\mu}(0) - \mu| \ll \mu$. To be able to apply the standard perturbation techniques for this system we must choose small gains: $K_\rho \ll |u|\mu$, $\sqrt{K_\mu} \ll \mu$, $K_\Delta \ll K_\mu\mu$.

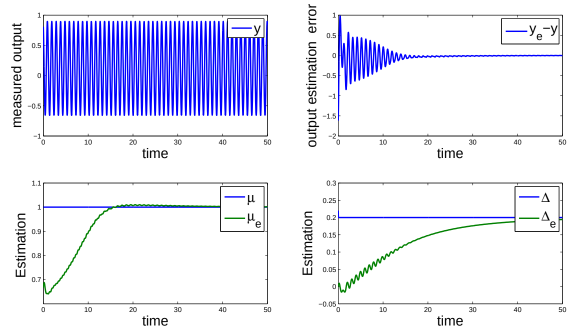

Fig. 1. Measured output, output error, estimations of the parameters μ and Δ without noise (y_e, Δ_e and μ_e stand for $\hat{y}$, $\hat{\Delta}$ and $\hat{\mu}$).

2.3 *Simulations*

In this subsection, we test the performance and the robustness of the above adaptive observer through numerical simulations. Note that the extension of the above algorithm will also be tested on a 3-level system in Subsection 5.2

We take for the initial conditions:

$$\rho_0 = \frac{1 + \cos\left(\frac{\pi}{5}\right)\sigma_x + \sin\left(\frac{\pi}{5}\right)\cos\left(\frac{\pi}{1.4}\right)\sigma_y + \sin\left(\frac{\pi}{5}\right)\sin\left(\frac{\pi}{1.4}\right)\sigma_z}{2}$$

$$\mu = 1\ , \quad \Delta = \frac{1}{5}\ , \quad \hat{\rho}_0 = \sigma_x\rho_0\sigma_x$$

We choose for the control u and the gains: $u = 1$, $K_\rho = 2\varepsilon|u|\mu$, $K_\mu = 2\varepsilon^2|u|\mu$ et $K_\Delta = 2\varepsilon^2|u|\mu$ with $\varepsilon = \frac{1}{5}$. The results are given by fig 1. As one can see, the adaptive observer ensures the convergence of the parameter estimators $\hat{\mu}$ and $\hat{\Delta}$ as well as the output error $\mathrm{Tr}(\sigma_z\hat{\rho}) - y$. Moreover, the convergence of the estimator $\hat{\mu}$ takes place at a faster rate than the estimator $\hat{\Delta}$.

In fig 2, the measured signals y and laser amplitude u were added white gaussian independent noises of amplitude 20% (output y) and 10% (control u). One observes the robustness of the estimation algorithm with respect to all these uncertainties.

3 Properties of the system and the observer

3.1 *Symmetries*

The system is invariant under a change of basis for the wave function $\Psi \mapsto U\Psi$ where $U \in SU(2)$. Indeed consider the transformation $\varpi = U\rho U^\dagger$, and $\zeta_x = U\sigma_x U^\dagger$, $\zeta_y = U\sigma_y U^\dagger$,

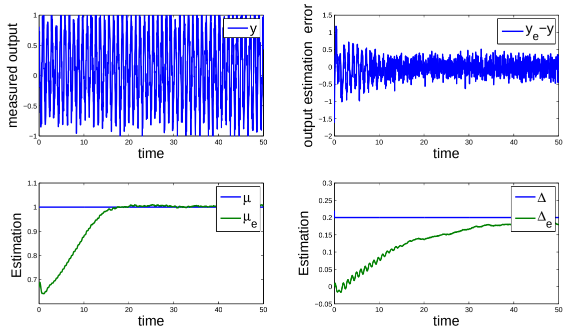

Fig. 2. Measured output, output error, estimations of the parameters μ and Δ with noisy measurement and control (y_e, Δ_e and μ_e stand for $\hat{y}$, $\hat{\Delta}$ and $\hat{\mu}$).

$\zeta_z = U\sigma_z U^\dagger$. With the new variables, the dynamics (2)-(5) writes

$$\frac{d}{dt}\varpi = -\imath[\frac{\Delta}{2}\zeta_z + \frac{u\mu}{2}\zeta_x, \varpi]$$
$$y = Tr(\zeta_z \varpi)$$

Δ and μ are unchanged by the transformation and we still have $\dot{\Delta} = \dot{\mu} = 0$. ζ_x, ζ_y, ζ_z verify the commutation relations of the Pauli matrices. Thus the system is invariant under the action of the transformation group $SU(2)$ (see definition 2 of Appendix A).

3.2 *Invariants, Invariant output, invariant vector fields*

Here, we explain the structure of the (symmetry-preserving) observer (6)-(7)-(8). In this subsection, we consider the reduced system (2)-(5)

$$\frac{d}{dt}\rho = -\imath\left[\frac{\Delta}{2}\sigma_z + \frac{u\mu}{2}\sigma_x, \rho\right]$$
$$y = Tr(\sigma_z\rho)$$

since the action of the group SU(2) on Δ and μ is trivial. To build observers which preserve the symmetries (i.e, are invariant under the action of $SU(2)$), we follow the method of [5] : find a) an invariant output error b) an invariant frame c) a complete set of scalar invariants d) use eq (A.4) to derive non-linear observers. Some basic definitions and results of this latter paper are recalled in the appendix A.

The method does not fully apply to the two-state quantum system under study, since the dimension of the group $SU(2)$ is 3 and the group acts non-trivially on ρ which is of dimension 2, and for any density matrix ρ the set of all $U \in SU(2)$ such that $U\rho U^\dagger = \rho$ (isotropy subgroup) is of dimension 1. It contradicts the full-rank assumptions on the group action (free action) of [5]. Nevertheless the method (although local) gives very useful guidelines for the design of (globally defined) non-linear observers.

Invariant output error:

The output $y = Tr(\sigma_z\rho)$ is a scalar. It is invariant under the group action (it has a physical meaning independent of the basis used to write the Schrödinger equation). Indeed for any $U \in SU(2)$ if we let $\varpi = U\rho U^\dagger$ and $\zeta_z = U\sigma_z U^\dagger$ we have $y = Tr(\zeta_z\varpi) = Tr(U\sigma_z\rho U^\dagger) = Tr(\sigma_z\rho)$. Thus an output error is (see the definition 5 of Appendix A) :

$$E(\hat{\rho}, \sigma_x, \sigma_y, \sigma_z, y) = \hat{y} - y = Tr(\sigma_z(\hat{\rho} - \rho))$$

Invariant vector fields:

The system (2) is invariant thus the second member of (2) is made of invariant vector fields (in the sense of the definition 3 of Appendix A). Inspiring by equation (2) let us take as invariant vector fields

$$(w_1(\rho), w_2(\rho), w_3(\rho)) = (-\imath[\sigma_x, \rho], -\imath[\sigma_y, \rho], -\imath[\sigma_z, \rho])$$

They are not an invariant frame (see def 4) since they are functionally dependant. Indeed the tangent space of the 2×2 projector matrices with trace 1 at any point is only of dimension 2. But they provide a global parameterization of the tangent bundle.

Scalar invariants:

A complete set of scalar invariants is a full rank function $(\hat{\rho}, \sigma_x, \sigma_y, \sigma_z) \mapsto I(\hat{\rho}, \sigma_x, \sigma_y, \sigma_z) \in \mathbb{R}^{n+m-r}$ which is invariant under the group action (where n is the dimension of the state space, m is the dimension on the inputs on which $(\psi_g)_{g\in G}$ act, and r is the dimension of the group G). Here $n = 2, m = 3$ and $r = 3$. Locally there are $n+m-r = 2+3-3 = 2$ independent scalar invariants (see [30]). Take :

$$I(\hat{\rho}, \sigma_x, \sigma_y, \sigma_z) = (Tr(\sigma_x\hat{\rho}), Tr(\sigma_y\hat{\rho}), Tr(\sigma_z\hat{\rho}))$$

It is a set of 3 functionally dependent global invariants since $Tr^2(\sigma_x\hat{\rho}) + Tr^2(\sigma_y\hat{\rho}) + Tr^2(\sigma_z\hat{\rho}) = 1$ (see section 3.3).

We are going to prove now that the observer (6) for the reduced system (2)-(5) corresponds indeed to (A.4) and thus is a symmetry-preserving observer.

3.3 *Geometrical interpretation with the Bloch sphere*

The Bloch sphere is a geometrical representation of the pure state space of a two-level quantum mechanical system. An important property is that any density matrix ρ can be written

$$\rho = \frac{1 + X\sigma_x + Y\sigma_y + Z\sigma_z}{2}, \quad \text{with } \zeta = \begin{pmatrix} X \\ Y \\ Z \end{pmatrix} \in \mathbb{S}^2$$

where 1 denotes the identity 2×2 matrix. We have $\mathrm{Tr}(\sigma_x\rho)=X, \mathrm{Tr}(\sigma_y\rho)=Y$ and $\mathrm{Tr}(\sigma_z\rho)=Z$. Note that, the coordinate Z in this section is the output y. The commutation operation $-\imath[\sigma_x,\rho]$ corresponds to the wedge product $(1,0,0)^T$ with $(X,Y,Z)^T$ (one has similar correspondences for other indices y and z). The dynamics of ζ (see eq (2)) is

$$\frac{d}{dt}\zeta=(\frac{u\mu}{2},0,\frac{\Delta}{2})^T\wedge\zeta \tag{9}$$

The symmetries of section 3.1 associated to the invariance under a change of basis correspond in the Bloch sphere representation to the invariance with respect to the choice of any orthonormal frame (invariance under the action of SO(3)). Physically, designing a symmetry-preserving observer amounts to compel the estimated system to be pure (i.e, $\hat{\zeta}\in\mathbb{S}^2$).

The correction term in (6) writes $-K_\rho(\hat{Z}-Z)[-\hat{X}\hat{Z}\sigma_x-\hat{Y}\hat{Z}\sigma_y+(1-\hat{Z}^2)\sigma_z]$ and corresponds in the Bloch sphere to $-K_\rho(\hat{Z}-Z)(\hat{Y},-\hat{X},0)^T\wedge\hat{\zeta}$, where $\hat{\zeta}=(\hat{X},\hat{Y},\hat{Z})^T$. It means that (6) can be written

$$\begin{aligned}\frac{d}{dt}\hat{\rho}=&-\imath\left[\frac{\hat{\Delta}}{2}\sigma_z+\frac{u\hat{\mu}}{2}\sigma_x,\hat{\rho}\right]\\&+\imath K_\rho(\mathrm{Tr}(\sigma_z\hat{\rho})-y)\ (\mathrm{Tr}(\sigma_y\hat{\rho})[\sigma_x,\hat{\rho}]-\mathrm{Tr}(\sigma_x\hat{\rho})[\sigma_y,\hat{\rho}])\end{aligned}$$

and thus the observer (6) for the reduced system (2)-(5) is of the form(A.4).

Heuristic of the invariant correction terms

This paragraph explains the geometrical motivations for the gains we chose. More rigorous proofs are given in the next section. Let $e_z=(0,0,1)^T$. We proved that (6) writes in the Bloch sphere :

$$\frac{d}{dt}\hat{\zeta}=\frac{1}{2}\begin{pmatrix}u\hat{\mu}\\0\\\hat{\Delta}\end{pmatrix}\times\hat{\zeta}-K_\rho(\hat{Z}-Z)\big(\hat{\zeta}\times e_z\big)\times\hat{\zeta} \tag{10}$$

Thus the correction term (6) is "mirrored" in the Bloch sphere representation by the projection of the gradient of $-\frac{K_\rho}{2}(\hat{Z}-Z)^2$ on the tangent space $T\mathbb{S}^2|_{\hat{\zeta}}$. It is a vector which is always pointing towards "north" (i.e, $Z=1$) if $\hat{Z}<Z$ and towards "south" (i.e, $Z=-1$) if not.

K_ρ is much larger than the gains K_μ and K_Δ (the ratio is of order ε). Let us now suppose that $\hat{\zeta}-\zeta\approx 0$. Using (9) and (10), we have $\frac{d}{dt}(\hat{Z}-Z)\approx\frac{u}{2}(\hat{\mu}-\mu)\hat{Y}$. Thus, if $\hat{\mu}>\mu$, the difference $\hat{Z}-Z$ tends to increase iff $\hat{Y}>0$. This explains the choice of (7) which writes in the Bloch sphere representation $\frac{d}{dt}\hat{\mu}=-uK_\mu\hat{Y}(\hat{Z}-Z)$. The design of (8) which writes $\frac{d}{dt}\hat{\Delta}=-uK_\Delta\hat{X}(\hat{Z}-Z)$ can be explained the same way assuming moreover that $\hat{\mu}-\mu\approx 0$ and noticing then $\frac{d^2}{dt^2}(\hat{Z}-Z)\approx\frac{u}{4}\mu(\hat{\Delta}-\Delta)\hat{X}$.

4 Convergence analysis

Theorem 1 *Consider the two-level system described by* (2)-(3)-(4)-(5) *where the amplitude of the laser u is constant. Assume $Tr(\sigma_x\rho_0)\neq -1,0,1$, $\mu\neq 0$ and $u\neq 0$. For any positive tuning parameters k_ρ,k_μ,k_Δ, there exists $\overline{\varepsilon}>0$ such that for any $\varepsilon\in]0,\overline{\varepsilon}]$, if*

$$\begin{aligned}&\Delta\leq\varepsilon|u\mu|,\quad K_\rho=2k_\rho\varepsilon|u|\mu,\\&K_\mu=2k_\mu\varepsilon^2\mu^2,\quad K_\Delta=2k_\Delta\varepsilon^2|u|\mu^2,\end{aligned} \tag{11}$$

the non-linear observer (6)-(7)-(8) *is locally convergent in the following sense: there exists $\delta>0$ such that if its initial condition* $(\hat{\rho}_0,\hat{\mu}_0,\hat{\Delta}_0)$ *satisfies*

$$|\hat{\rho}_0-\rho_0|\leq\delta,\ |\hat{\mu}_0-\mu|\leq\delta\varepsilon|\mu|,\ |\hat{\Delta}_0-\Delta|\leq\delta\varepsilon|u\mu|,$$

$(\hat{\rho},\hat{\mu},\hat{\Delta})$ *converges towards* (ρ,μ,Δ) *when* $t\to+\infty$.

Sketch of the proof. First we make a standard time-varying transformation to apply the averaging method called rotating wave approximation. With the gains given by the theorem the approximation leads to an approximation of the system to order ε being valid for a time $O(1/\varepsilon)$: system (19)-(20)-(21)-(22). Using the coordinates in the Bloch sphere, we derive a Lyapunov function for this first-order system proving the local and exponential convergence of the state error $\hat{\rho}-\rho$ and the parameter error $\hat{\mu}-\mu$ via Lasalle's principle. This first approximation being insufficient to prove the convergence of $\hat{\Delta}$ to Δ, an approximation to second order terms in ε is needed: system (26). This final system has a *triangular* structure, and the convergence of $\hat{\rho}-\rho$ and $\hat{\mu}-\mu$ to 0 (already proved) imply immediately the convergence of $\hat{\Delta}$. We conclude using standard averaging arguments relying on the exponential convergence of the second-order approximated system.

PROOF. The proof of the theorem relies on a standard averaging method called rotating wave approximation. To apply it one writes the system in the (standard) interaction frame, i.e, one makes the time dependant change of variables:

$$\rho=e^{-\imath\frac{u\mu t\sigma_x}{2}}\xi e^{\imath\frac{u\mu t\sigma_x}{2}},\qquad\hat{\rho}=e^{-\imath\frac{u\mu t\sigma_x}{2}}\hat{\xi}e^{\imath\frac{u\mu t\sigma_x}{2}}.$$

We have thus

$$\frac{d}{dt}\xi=[\imath\frac{u\mu\sigma_x}{2},\xi]+e^{\imath\frac{u\mu t\sigma_x}{2}}(\frac{d}{dt}\rho)e^{-\imath\frac{u\mu t\sigma_x}{2}}$$

as well as a similar relation for $\frac{d}{dt}\hat{\xi}$. Throughout the section, we are going to use the two following useful features: for any scalar a we have

$$e^{\imath a\sigma_x} = \cos a + \imath \sin a\ \sigma_x \quad \text{thus} \quad e^{\imath a\sigma_x}\sigma_y = \sigma_y e^{-\imath a\sigma_x} \tag{12}$$

and thus for any 2×2 matrix M

$$\begin{aligned} e^{\imath\frac{a}{2}\sigma_x}[\sigma_y, M]e^{-\imath\frac{a}{2}\sigma_x} &= [e^{\imath a\sigma_x}\sigma_y, e^{\imath\frac{a}{2}\sigma_x}Me^{-\imath\frac{a}{2}\sigma_x}] \\ e^{\imath\frac{a}{2}\sigma_x}[\sigma_x, M]e^{-\imath\frac{a}{2}\sigma_x} &= [\sigma_x, e^{\imath\frac{a}{2}\sigma_x}Me^{-\imath\frac{a}{2}\sigma_x}] \end{aligned} \tag{13}$$

Both preceding formulas hold for any circular permutation of the indices x, y and z. Thus

$$\frac{d}{dt}\xi = -\imath \left[\frac{\Delta}{2}e^{\imath u\mu t\sigma_x}\sigma_z, \xi\right] \tag{14}$$

which demonstrates the reason to use the interaction picture (the free dynamics being removed). Let

$$\begin{aligned} &\Delta = \varepsilon|u\mu|\chi, \quad \hat{\Delta} = \varepsilon|u\mu|\hat{\chi}, \quad \hat{\mu} - \mu = \varepsilon\hat{\vartheta}\ \mu, \\ &K_\rho = 2k_\rho\varepsilon|u|\mu, \quad K_\mu = 2k_\mu\varepsilon^2\mu^2, \quad K_\Delta = 2k_\Delta\varepsilon^2|u|\mu^2. \end{aligned} \tag{15}$$

Still using (12)-(13), the observer in the interaction frame reads (assuming u is real for simplicity's sake, i.e. $u = |u|$):

$$\begin{aligned} &\frac{d}{dt}\hat{\xi} = -\imath\varepsilon|u\mu|\left[\frac{\hat{\chi}}{2}e^{\imath u\mu t\sigma_x}\sigma_z + \frac{\hat{\vartheta}}{2}\sigma_x, \hat{\xi}\right] \\ &-2\varepsilon|u\mu|k_\rho \mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z(\hat{\xi}-\xi)\right)\times \\ &\qquad\left(e^{\imath u\mu t\sigma_x}\sigma_z\hat{\xi} + \hat{\xi}e^{\imath u\mu t\sigma_x}\sigma_z - 2\mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z\hat{\xi}\right)\hat{\xi}\right) \\ &\frac{d}{dt}\hat{\vartheta} = -2\varepsilon|u\mu|k_\mu \mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_y\hat{\xi}\right)\ \mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z(\hat{\xi}-\xi)\right) \\ &\frac{d}{dt}\hat{\chi} = -2\varepsilon|u\mu|k_\Delta \mathrm{Tr}\left(\sigma_x\hat{\xi}\right)\ \mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z(\hat{\xi}-\xi)\right). \end{aligned} \tag{16}$$

First-order secular approximation

The integration of terms of the form $\exp(iku\mu t)$, $k\in\mathbb{N}^*$ over the time t yields terms of small amplitude and oscillating with high frequency and zero mean. The secular approximation consists in neglecting the terms rotating with high frequencies $u\mu$ and $2u\mu$, by averaging their influence on the evolution of ρ (see, e.g., [13] for a physicist point of view on the standard rotating wave approximation or [1] for a more formal exposure).The true dynamics consists of (fast) small oscillations around the (slowly-varying) solution of the averaged system. In order to compute the averaged system we use (12) to prove for any scalar a

$$e^{\imath a\sigma_x}\sigma_z = \cos(a)\sigma_z + \sin(a)\sigma_y \tag{17}$$
$$e^{\imath a\sigma_x}\sigma_y = \cos(a)\sigma_y - \sin(a)\sigma_z. \tag{18}$$

and thus (for instance)

$$\begin{aligned} &\mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z(\hat{\xi}-\xi)\right)e^{\imath u\mu t\sigma_x}\sigma_z\hat{\xi} \\ &= \cos^2(u\mu t)\mathrm{Tr}\left(\sigma_z(\hat{\xi}-\xi)\right)\sigma_z\hat{\xi} \\ &\quad + \sin^2(u\mu t)\mathrm{Tr}\left(\sigma_y(\hat{\xi}-\xi)\right)\sigma_y\hat{\xi} \\ &\quad + \sin(u\mu t)\cos(u\mu t)\times \\ &\qquad\left(\mathrm{Tr}\left(\sigma_z(\hat{\xi}-\xi)\right)\sigma_y\hat{\xi} + \mathrm{Tr}\left(\sigma_y(\hat{\xi}-\xi)\right)\sigma_z\hat{\xi}\right). \end{aligned}$$

Computing all the other terms of (16) the same way, and taking the time average over a period $\pi/(u\mu)$ (assuming that ξ, $\hat{\xi}$, $\hat{\Delta}$, $\hat{\mu}$ are constant over a period), we get the following autonomous system for the averaged system/observer:

$$\frac{d}{dt}\xi = 0\ , \tag{19}$$

$$\begin{aligned} \frac{d}{dt}\hat{\xi} = &-\imath\ \varepsilon|u\mu|\left[\frac{\hat{\vartheta}}{2}\sigma_x, \hat{\xi}\right] \\ &-\varepsilon|u\mu|k_\rho \mathrm{Tr}\left(\sigma_y(\hat{\xi}-\xi)\right)\left(\sigma_y\hat{\xi} + \hat{\xi}\sigma_y - 2\mathrm{Tr}\left(\sigma_y\hat{\xi}\right)\hat{\xi}\right) \\ &-\varepsilon|u\mu|k_\rho \mathrm{Tr}\left(\sigma_z(\hat{\xi}-\xi)\right)\left(\sigma_z\hat{\xi} + \hat{\xi}\sigma_z - 2\mathrm{Tr}\left(\sigma_z\hat{\xi}\right)\hat{\xi}\right), \end{aligned} \tag{20}$$

$$\begin{aligned} \frac{d}{dt}\hat{\vartheta} = -\varepsilon|u\mu|k_\mu[&\mathrm{Tr}\left(\sigma_y\hat{\xi}\right)\mathrm{Tr}\left(\sigma_z(\hat{\xi}-\xi)\right) \\ &-\mathrm{Tr}\left(\sigma_z\hat{\xi}\right)\mathrm{Tr}\left(\sigma_y(\hat{\xi}-\xi)\right)], \end{aligned} \tag{21}$$

$$\frac{d}{dt}\hat{\chi} = 0. \tag{22}$$

At this point of the proof, we show that the above system (19)-(20)-(21) is locally exponentially stable. This means that for any choice of $k_\rho, k_\mu > 0$, the estimates $\hat{\xi}-\xi$ and $\hat{\vartheta}$ both converge locally exponentially to 0.

Lyapunov function for the first-order secular approximation

We derive here a Lyapunov function to study the behaviour of the above first-order approximated system. The density matrices are well suited for computing the system in the interaction frame, using intensively (12)-(13), and for averaging. Such computations using rotation matrices and coordinates on the Bloch sphere would demand many more calculations. But it is illuminating (although not necessary) to write the averaged system in the Bloch sphere. As in section 3.3 let $\xi = \frac{1+X\sigma_x+Y\sigma_y+Z\sigma_z}{2}$ and $\hat{\xi} = \frac{1+\hat{X}\sigma_x+\hat{Y}\sigma_y+\hat{Z}\sigma_z}{2}$. We have $X^2+Y^2+Z^2 = \hat{X}^2+\hat{Y}^2+\hat{Z}^2 = 1$. Note that it is due to the fact that the observer preserves the geometry of the system. The averaged system writes

$$
\begin{aligned}
&\frac{d}{dt}X = \frac{d}{dt}Y = \frac{d}{dt}Z = 0\\
&\frac{d}{dt}\hat{X} = \varepsilon|u\mu|k_\rho\big([(\hat{Y}-Y)\hat{X}\hat{Y} + (\hat{Z}-Z)\hat{X}\hat{Z}]\big)\\
&\frac{d}{dt}\hat{Y} = \varepsilon|u\mu|\big(-\frac{\hat{\vartheta}}{2}\hat{Z} - k_\rho[(\hat{Y}-Y)(\hat{X}^2+\hat{Z}^2)] + k_\rho[(\hat{Z}-Z)\hat{Z}\hat{Y}]\big)\\
&\frac{d}{dt}\hat{Z} = \varepsilon|u\mu|\big(\frac{\hat{\vartheta}}{2}\hat{Y} + k_\rho[(\hat{Y}-Y)\hat{Z}\hat{Y}] - k_\rho[(\hat{Z}-Z)(\hat{X}^2+\hat{Y}^2)]\big)\\
&\frac{d}{dt}\hat{\vartheta} = -\varepsilon|u\mu|k_\mu[\hat{Y}(\hat{Z}-Z) - \hat{Z}(\hat{Y}-Y)]
\end{aligned}
$$

We consider the Lyapunov function

$$V = (\hat{Y}-Y)^2 + (\hat{Z}-Z)^2 + \frac{1}{2k_\mu}\hat{\vartheta}^2$$

Using $\hat{X}^2+\hat{Z}^2 = 1-\hat{Y}^2$, we have

$$
\begin{aligned}
\frac{dV}{dt} =& 2\varepsilon|u\mu|k_\rho\big(-(\hat{Y}-Y)^2(1-\hat{Y}^2) + 2(\hat{Y}-Y)(\hat{Z}-Z)\hat{Z}\hat{Y}\\
&-(\hat{Z}-Z)^2(1-\hat{Z}^2)\big)\\
=& -2\varepsilon|u\mu|k_\rho\left((\hat{Y}-Y)\hat{Z} - (\hat{Z}-Z)\hat{Y}\right)^2\\
&-2\varepsilon|u\mu|k_\rho\hat{X}^2\left((\hat{Y}-Y)^2 + (\hat{Z}-Z)^2\right) \quad \leq 0.
\end{aligned}
$$

Since ξ and $\hat{\xi}$ evolve on a compact manifold and V is infinite when $\hat{\vartheta}$ tends to infinity, one can apply the LaSalle invariance principle. Simple computations show that the Lasalle's invariant set reduces to 4 equilibrium points for $(\hat{X},\hat{Y},\hat{Z},\hat{\vartheta})$:

$$\left(0, \pm\frac{Y}{\sqrt{Y^2+Z^2}}, \pm\frac{Z}{\sqrt{Y^2+Z^2}}, 0\right), \quad (\pm X, Y, Z, 0)$$

Computing their Lyapunov exponents, the first two are unstable and the last two, and in particular $(X,Y,Z,0)$, are hyperbolically stable. Notice that in any case $\hat{\vartheta}$ converges to 0. So we have proved that the steady-state $(\hat{\xi},\hat{\vartheta}) = (\xi,0)$ of the averaged system (19)-(20)-(21) is exponentially stable.

Second-order secular approximation

Since the first-order secular (non-oscillating) terms vanish when computing $\frac{d}{dt}\xi$ and $\frac{d}{dt}\hat{\chi}$, we have to consider the second-order secular approximation to prove convergence. We apply the Kapitsa shortcut method described in e.g, [19]. ξ obeys a differential equation with a high frequency source term $\frac{d}{dt}\xi = \varepsilon u\mu f(\xi, u\mu t)$ where $\varepsilon \ll 1$. We proved that the mean of ξ over a period is constant. Integrating high frequency terms yields high frequency terms with same frequency and smaller amplitude: we thus seek a solution of the type

$$\xi = \zeta + \varepsilon g_1(\zeta, u\mu t) + \varepsilon^2 g_2(\zeta, u\mu t) + \cdots$$

where ζ is the mean of ξ over a period (recall $\frac{d}{dt}\zeta = 0 + O(\varepsilon)$). Let us compute the second-order term, $g_1(\zeta, u\mu t)$, and neglect the third-order terms. On one hand, we have:

$$\frac{d}{dt}\xi = 0 + \varepsilon u\mu\ \partial_2 g_1(\zeta, u\mu t)$$

using the partial derivative of g_1 with respect to its second variable. But using (14) and neglecting third order terms:

$$\frac{d}{dt}\xi = -\imath\left[\frac{\varepsilon|u\mu|\chi}{2}e^{\imath u\mu t\sigma_x}\sigma_z, \zeta + \varepsilon g_1(\zeta, u\mu t)\right]. \tag{23}$$

Gathering the two latter equations

$$\partial_2 g_1(\zeta, u\mu t) = -\imath[\frac{\chi}{2}e^{\imath u\mu t\sigma_x}\sigma_z, \zeta] + O(\varepsilon).$$

Integrating with respect to time t the last equation yields:

$$g_1(\zeta, u\mu t) = \imath[\frac{\chi}{2}e^{\imath u\mu t\sigma_x}\sigma_y, \xi] + O(\varepsilon).$$

Thus (23) can be re-written as

$$
\begin{aligned}
\frac{d}{dt}\xi &= -\imath\varepsilon|u\mu|[\frac{\chi}{2}e^{\imath u\mu t\sigma_x}\sigma_z, \zeta + \varepsilon g_1(\zeta,t) + O(\varepsilon^2)]\\
&= -\imath\varepsilon|u\mu|[\frac{\chi}{2}e^{\imath u\mu t\sigma_x}\sigma_z, \zeta]\\
&\quad + \varepsilon^2|u\mu|\frac{\chi^2}{4}[e^{\imath u\mu t\sigma_x}\sigma_z, [e^{\imath u\mu t\sigma_x}\sigma_y, \zeta]] + O(\varepsilon^3)
\end{aligned}
$$

Now let us compute the temporal mean (over a period) and only keep the secular terms. We apply (17) and (18). The Jacobi identity yields

$$\frac{d}{dt}\xi = -\imath\varepsilon^2|u\mu|\frac{\chi^2}{2}[\sigma_x, \xi] + \cdots$$

where we have not written the oscillating terms of 0 mean nor the terms of order $O(\varepsilon^3)$. Note that, here, we find the second-order term $\frac{\Delta^2}{2u\mu}$ corresponding to the standard Bloch-Siegert shift.

We also need to write $\hat{\chi}$ up to second order terms since it has constant mean. We go back now to the exact equations (16). We have

$$\frac{d}{dt}\hat{\chi} = -2\varepsilon|u\mu|k_\Delta \mathrm{Tr}\left(\sigma_x\hat{\xi}\right)\ \mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z(\hat{\xi}-\xi)\right) \tag{24}$$

Up to the second order, the secular terms of (24) can be calculated as the sum of two parts: 1. replace $\hat{\xi}-\xi$ by its $u\mu$-frequency part in order to compute the secular terms of $\mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z(\hat{\xi}-\xi)\right)$; then multiply them by

$-2\varepsilon|u\mu|k_\Delta \mathrm{Tr}\left(\sigma_x\hat{\xi}\right)$; 2. replace $\hat{\xi}$ in $\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)$ by its $u\mu$-frequency terms, leaving $\hat{\xi}-\xi$ untouched and calculate the secular terms of $\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)\ \mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z(\hat{\xi}-\xi)\right)$. Note that, according to (16), ξ and $\hat{\xi}$ up to second order have an oscillating part of frequency $u\mu$ and another one of frequency $2u\mu$. But only the terms of frequency $u\mu$ in ξ and $\hat{\xi}$ can have a secular effect when following the two steps 1. and 2.

The $u\mu$-frequency part of $\hat{\xi}$ is due to the integration of $-\imath\varepsilon|u\mu|\ \left[\frac{\hat{\chi}}{2}e^{\imath u\mu t\sigma_x}\sigma_z,\hat{\xi}\right]$ and so is for ξ. Thus

$$
\begin{aligned}
\hat{\xi} &= \hat{\zeta}+\varepsilon\frac{\imath\hat{\chi}}{2}\left[e^{\imath u\mu t\sigma_x}\sigma_y,\hat{\xi}\right]+\cdots\\
\xi &= \zeta+\varepsilon\frac{\imath\chi}{2}\left[e^{\imath u\mu t\sigma_x}\sigma_y,\xi\right]+\cdots
\end{aligned}
\tag{25}
$$

where $\hat{\zeta}$ (resp ζ) is a solution of the averaged equation for $\hat{\xi}$ (resp ξ) and the non-written terms are either $2u\mu$-frequency terms or are of order $O(\varepsilon^2)$. So 1. on one hand, we have

$$
\mathrm{Tr}\left(e^{\imath u\mu t\sigma_x}\sigma_z(\hat{\xi}-\xi)\right)=\varepsilon\left(\hat{\chi}\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)-\chi\mathrm{Tr}\left(\sigma_x\xi\right)\right)+\cdots
$$

where we have not written the oscillating terms of 0 mean. Here, we have applied (25) and the following relation which can be proven using (12) and (13):

$$
\frac{\hat{\chi}}{2}e^{\imath u\mu t\sigma_x}\sigma_z[e^{\imath u\mu t\sigma_x}\sigma_y,\hat{\xi}]=\frac{\hat{\chi}}{2}\left(\sigma_z[\sigma_y,\hat{\xi}]-\imath\sigma_y[\sigma_y,\hat{\xi}]\right)+\cdots
$$

2. on the other hand, for the $u\mu$-frequency part of $\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)$, we use (25), and using (12) we have:

$$
\mathrm{Tr}\left(\sigma_x[e^{\imath u\mu t\sigma_x}\sigma_y,\hat{\xi}]\right)=2\imath\mathrm{Tr}\left(\cos u\mu t\ \sigma_z\hat{\xi}+\sin u\mu t\ \sigma_y\hat{\xi}\right).
$$

We can then write the sine and cosine as sums of exponentials and the secular terms give the last term our final intermediary result which is that, up to the second order

$$
\begin{aligned}
\frac{d}{dt}\hat{\chi} = & -2\varepsilon^2|u\mu|k_\Delta\left(\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)^2\hat{\chi}-\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)\mathrm{Tr}\left(\sigma_x\xi\right)\chi\right)\\
& +\varepsilon^2|u\mu|k_\Delta\hat{\chi}[\mathrm{Tr}\left(\sigma_y\hat{\xi}\right)\mathrm{Tr}\left(\sigma_y(\hat{\xi}-\xi)\right)\\
& -\mathrm{Tr}\left(\sigma_z\hat{\xi}\right)\mathrm{Tr}\left(\sigma_z(\hat{\xi}-\xi)\right)].
\end{aligned}
$$

We have obtained the following, locally convergent, triangular system:

$$
\begin{aligned}
\frac{d}{dt}\xi &\overset{\text{order }2}{=} -\imath\varepsilon^2|u\mu|\frac{\chi^2}{2}[\sigma_x,\xi] \qquad (26)\\
\frac{d}{dt}\hat{\xi} &\overset{\text{order }1}{=} -\imath\ \varepsilon|u\mu|\left[\frac{\hat{\vartheta}}{2}\sigma_x,\hat{\xi}\right]\\
&\quad -\varepsilon|u\mu|k_\rho\mathrm{Tr}\left(\sigma_y(\hat{\xi}-\xi)\right)\left(\sigma_y\hat{\xi}+\hat{\xi}\sigma_y-2\mathrm{Tr}\left(\sigma_y\hat{\xi}\right)\hat{\xi}\right)\\
&\quad -\varepsilon|u\mu|k_\rho\mathrm{Tr}\left(\sigma_z(\hat{\xi}-\xi)\right)\left(\sigma_z\hat{\xi}+\hat{\xi}\sigma_z-2\mathrm{Tr}\left(\sigma_z\hat{\xi}\right)\hat{\xi}\right),\\
\frac{d}{dt}\hat{\vartheta} &\overset{\text{order }1}{=} -\varepsilon|u\mu|k_\mu[\mathrm{Tr}\left(\sigma_y\hat{\xi}\right)\mathrm{Tr}\left(\sigma_z(\hat{\xi}-\xi)\right)\\
&\quad -\mathrm{Tr}\left(\sigma_z\hat{\xi}\right)\mathrm{Tr}\left(\sigma_y(\hat{\xi}-\xi)\right)],
\end{aligned}
$$

$$
\begin{aligned}
\frac{d}{dt}\hat{\chi} &\overset{\text{order }2}{=} -2\varepsilon^2|u\mu|k_\Delta\left(\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)^2\hat{\chi}-\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)\mathrm{Tr}\left(\sigma_x\xi\right)\chi\right)\\
&\quad +\varepsilon^2|u\mu|k_\Delta\hat{\chi}[\mathrm{Tr}\left(\sigma_y\hat{\xi}\right)\mathrm{Tr}\left(\sigma_y(\hat{\xi}-\xi)\right) \qquad (27)\\
&\quad -\mathrm{Tr}\left(\sigma_z\hat{\xi}\right)\mathrm{Tr}\left(\sigma_z(\hat{\xi}-\xi)\right)].
\end{aligned}
$$

In the first step of the proof (first-order secular approximation), we proved that $\hat{\vartheta}$ and $\hat{\xi}-\xi_0$ converge (locally) to 0 for any $k_\rho,k_\mu>0$ independent of $\hat{\chi}$. The last equation of (26) shows that once $\hat{\xi}-\xi$ is close to 0, since we supposed $\mathrm{Tr}\left(\sigma_x\xi_0\right)\neq -1,0,1$, the dominant term multiplying $\hat{\chi}$ is $-\mathrm{Tr}\left(\sigma_x\hat{\xi}\right)^2$, and $\hat{\chi}-\chi$ converges to 0 for $k_\Delta>0$. This implies that the point $(\xi_0,0,\chi)$ is a hyperbolic, asymptotically stable, fixed point of the averaged dynamics (26).

We can, now, apply the classical averaging theorem (see e.g. Theorem 4.1.1, page 168 of [12]). Note that, even though the cited theorem deals with the first order approximations, the whole analysis permitting to pass from the secular approximation in bounded time horizon to an asymptotic result can be adapted to the situations of higher orders. Indeed, this analysis is only based on the application of the Poincaré map and the fact that in a bounded time horizon (of order $1/\varepsilon$) the averaged dynamics provide an $O(\varepsilon)$-estimate of the real dynamics.

Therefore, there exists a unique hyperbolic periodic orbit of the real system (6)-(7)-(8) of the form $\gamma_\varepsilon(t)=(\xi_0,0,\chi)+O(\varepsilon)$ which is also asymptotically stable. We conclude noting that $(\xi,0,\chi)$ actually provides such a periodic solution and therefore it is this asymptotically stable periodic orbit. □

5 Extension: multilevel case

In this section, we extend the above identification algorithm to the general case of a multi-level system. We will assume that we exactly know the system's energy levels and therefore that the system in the free Hamiltonian's rotating frame admits a zero detuning. The goal is therefore to identify the atom-laser coupling parameters. In the simulations, we will, however, see that the provided algorithm is in fact robust with respect to small uncertainties in the system's spectrum and that a small detuning will not affect the convergence of the algorithm.

Before going through this extension, we need to address the non-trivial identifiability problem for the multi-level case. The Appendix B (based on the result of [21]) provides sufficient assumptions ensuring the identifiability.

5.1 Observer design

Consider the N-level system, $(|j\rangle)_{j=1}^N$, described by the density matrix ρ obeying the following dynamics (we assume here the assumptions **A1**,**A2** and **A3** of the Appendix B to be satisfied):

$$\left\{ \frac{d}{dt}\rho = -i[H + u(t)\mu, \rho] \right. \tag{28}$$

where

- $H = \sum_{j=1}^N \omega_j |j\rangle\langle j|$ is the free Hamiltonian with ω_j real and satisfying $|\omega_l - \omega_k| \neq |\omega_{l'} - \omega_{k'}|$ for any distinct couples (l,k) and (l',k');
- $\mu = \sum_{1\leq l<k\leq N} \mu_{lk} (|k\rangle\langle l| + |l\rangle\langle k|)$ where μ_{lk} are the parameters to identify;
- the electromagnetic field is represented by the scalar input $u(t) \in \mathbb{R}$.

We assume that

$$y_j(t) = \mathrm{Tr}(P_j\rho(t)), \quad P_j = |j\rangle\langle j|, \quad j = 1,2,...,N$$

are the measured outputs. The goal is to estimate the coefficient μ_{lk}, the ω_j's being known.

We set $u(t) = \sum_{1\leq l<k\leq N} A_{lk}\cos(\omega_{lk}t)$ where $\omega_{lk} = \omega_l - \omega_k$ and A_{lk} is a constant amplitude.

Consider the Gellman matrices, i.e., "Pauli matrices" associated to the transition between l and k :

$$\begin{aligned} \sigma_x^{lk} &= |l\rangle\langle k| + |k\rangle\langle l| \\ \sigma_y^{lk} &= -i|l\rangle\langle k| + i|k\rangle\langle l| \\ \sigma_z^{lk} &= P_l - P_k = |l\rangle\langle l| - |k\rangle\langle k| \\ I^{lk} &= P_l + P_k = |l\rangle\langle l| + |k\rangle\langle k|. \end{aligned}$$

For each $l \neq k$, we have the usual relations:

$$(\sigma_x^{lk})^2 = I^{lk}, \quad \sigma_x^{lk}\sigma_y^{lk} = i\sigma_z^{lk}, \quad \ldots$$

For each j and $k \neq j$, $P_j = \frac{I^{jk}+\sigma_z^{jk}}{2}$. In the sequel, we use the shortcut notation $\sum_{lk}$ that stands for $\sum_{1\leq l<k\leq N}$. Thus we have

$$u = \sum_{lk} A_{lk}\cos(\omega_{lk}t), \quad \mu = \sum_{lk} \mu_{lk}\sigma_x^{lk}.$$

Thus (28) reads:

$$\frac{d}{dt}\rho = -i[H,\rho] - i\sum_{lk}\sum_{l'k'} A_{l'k'}\mu_{lk}\cos(\omega_{l'k'}t)[\sigma_x^{lk},\rho]$$

In the interaction frame, $\xi = e^{iHt}\rho e^{-iHt}$, we have

$$\frac{d}{dt}\xi = -i\sum_{lk}\sum_{l'k'} A_{l'k'}\mu_{lk}\cos(\omega_{l'k'}t)\left[e^{iHt}\sigma_x^{lk}e^{-iHt},\xi\right],$$

the measurement outputs being given by $y_j = \mathrm{Tr}(P_j\xi)$ since $[P_j,H] = 0$.

Simple computations show

$$e^{iHt}\sigma_x^{lk}e^{-iHt} = e^{i\omega_{lk}t\sigma_z^{lk}}\sigma_x^{lk} = \cos(\omega_{lk}t)\sigma_x^{lk} - \sin(\omega_{lk}t)\sigma_y^{lk}$$

and

$$e^{iHt}\sigma_y^{lk}e^{-iHt} = e^{i\omega_{lk}t\sigma_z^{lk}}\sigma_y^{lk} = \sin(\omega_{lk}t)\sigma_x^{lk} + \cos(\omega_{lk}t)\sigma_y^{lk}$$

For $(l,k) \neq (l',k')$, $|\omega_{lk}| \neq |\omega_{l'k'}|$. Thus resonant terms come only from $(l,k) = (l',k')$. Under the "rotating wave approximation", the above equation reads:

$$\frac{d}{dt}\xi = -i\sum_{lk}\frac{A_{lk}\mu_{lk}}{2}\left[\sigma_x^{lk},\xi\right]. \tag{29}$$

Note that when our knowledge of the energy levels, $(\omega_j)_{j=1}^N$, is uncertain, a detuning term has to be added to the dynamics:

$$\frac{d}{dt}\xi = -i\delta H - i\sum_{lk}\frac{A_{lk}\mu_{lk}}{2}\left[\sigma_x^{lk},\xi\right], \quad \delta H = \mathrm{diag}(\delta\omega_j). \tag{30}$$

Similarly to the 2-level case, we propose the following observer-based estimator:

$$\left\{ \begin{aligned} &\frac{d}{dt}\hat{\xi} = -i\sum_{lk}\frac{A_{lk}\hat{\mu}_{lk}}{2}\left[\sigma_x^{lk},\hat{\xi}\right] \\ &+\Gamma\sum_{j=1}^N \mathrm{Tr}\left(P_j(\xi-\hat{\xi})\right)\left(P_j\hat{\xi}+\hat{\xi}P_j - 2\mathrm{Tr}\left(P_j\hat{\xi}\right)\hat{\xi}\right) \\ &\frac{d}{dt}\hat{\mu}_{lk} = \frac{\gamma_{lk}A_{lk}}{2}\mathrm{Tr}\left(\sigma_y^{lk}\hat{\xi}\right)\mathrm{Tr}\left(\sigma_z^{lk}(\xi-\hat{\xi})\right), \end{aligned} \right. \tag{31}$$

noting that

$$\mathrm{Tr}\left(\sigma_z^{lk}(\xi-\hat{\xi})\right) = (y_l - \mathrm{Tr}\left(P_l\hat{\xi}\right)) - (y_k - \mathrm{Tr}\left(P_k\hat{\xi}\right)).$$

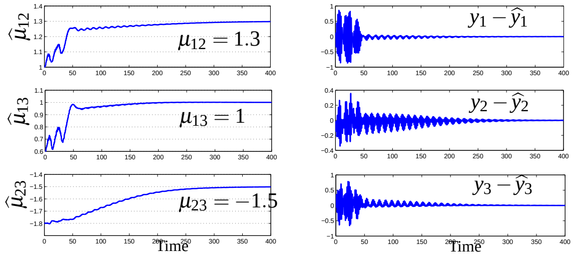

Fig. 3. Estimations of μ_{lk} and measurement output errors.

5.2 Numerical simulations

In this subsection, we check out the performance of the generalized estimator on a 3-level test case. The atom-laser coupling parameters, to be identified, are given as follows:

$$\mu = (\mu_{lk}) = \begin{pmatrix} 0 & 1.3 & 1 \\ 1.3 & 0 & -1.5 \\ 1 & -1.5 & 0 \end{pmatrix}$$

noting that by assumption **A3** of Appendix B, the diagonal part of μ must vanish. We consider the initial state

$$\rho_0 = \Psi_0 \Psi_0^*, \qquad \Psi_0 = \frac{1}{\sqrt{30}}(1,2,5)^T,$$

and the control amplitudes,

$$A_{lk} = .1, \qquad l \neq k.$$

Furthermore, we initialize the estimator as follows:

$$\begin{cases} \widehat{\mu} = (\widehat{\mu}_{lk}) = \begin{pmatrix} 0 & 1 & .6 \\ 1 & 0 & -1.8 \\ .6 & -1.8 & 0 \end{pmatrix} \\ \widehat{\rho}_0 = \widehat{\Psi}_0 \widehat{\Psi}_0^*, \qquad \widehat{\Psi}_0 = \frac{1}{\sqrt{14}}(1,2,3)^T. \end{cases}$$

In a first simulation, we consider an exact situation where no detuning, no noise in the measurement and no noise in the control amplitude are admitted. The simulations of Figure 3, then, correspond to the design parameters

$$\Gamma = 1, \qquad \gamma_{lk} = 0.1 \quad l \neq k.$$

In a second simulation (Figure 4), we consider a detuning of the form

$$\delta H = \mathrm{diag}(0, .01, .015),$$

an additive gaussian noise of 10% to the measurement outputs and a gaussian noise of 5% to the control amplitude.

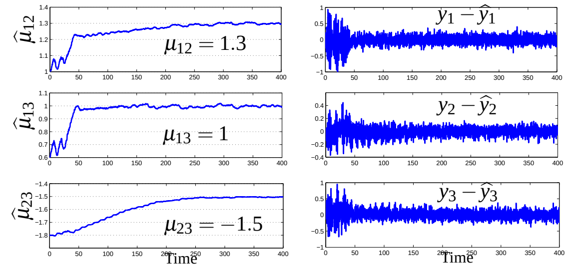

Fig. 4. Estimations of μ_{lk} and measurement output errors.

5.3 Convergence analysis

Consider the following function

$$V = \frac{1}{2}\sum_{n=1}^{N} \mathrm{Tr}\left(P_n(\hat{\xi} - \xi)\right)^2 + \sum_{lk} \frac{2(\hat{\mu}_{lk} - \mu_{lk})^2}{\gamma_{lk}}. \qquad (32)$$

One can easily see that

$$\frac{dV}{dt} = \sum_{lk} \frac{A_{lk}\mu_{lk}}{2} \mathrm{Tr}\left(\sigma_z^{lk}(\xi - \hat{\xi})\right) \mathrm{Tr}\left(\sigma_y^{lk}(\xi - \hat{\xi})\right)$$
$$- 2\Gamma \sum_{l}\sum_{k<l} \mathrm{Tr}\left(P_k\hat{\xi}\right) \mathrm{Tr}\left(P_l\hat{\xi}\right) \mathrm{Tr}\left(\sigma_z^{kl}(\xi - \hat{\xi})\right)^2. \qquad (33)$$

While the second term in (33) is obviously negative, the first term has no reason to be negative. However, we will show by a formal argument that (considering some appropriate assumption concerning the Rabi frequencies) this term can be averaged to zero and thus can be neglected.

In this aim, consider the real effective Hamitonian:

$$H_{eff} = \sum_{lk} \frac{A_{lk}\mu_{lk}}{2} \sigma_x^{lk},$$

and diagonalize it as follows:

$$H_{eff} = E^\dagger \Omega E, \quad \Omega = diag(\Omega_1, \ldots, \Omega_N), \quad E_{lk} \in \mathbb{R} \quad \forall l,k.$$

where $\{\Omega_j\}_{j=1}^N$ are Rabi frequencies of the system. From now on, we will assume that these Rabi frequencies are non-degenerate ($\Omega_m \neq \Omega_n$ for $m \neq n$) and moreover that $\Gamma \ll \Delta_\Omega$ and $\gamma_{lk} \ll \Delta_\Omega$, where $\Delta_\Omega = \max_{m\neq n} |\Omega_m - \Omega_n|$.

Now, in analogy with the 2-level case, consider the unitary transformation

$$\zeta = U^\dagger E \xi E^\dagger U, \quad \hat{\zeta} = U^\dagger E \hat{\xi} E^\dagger U,$$

where $U(t) = \exp(-it\Omega)$. Under such a transformation ζ is trivially constant $\zeta = E\xi_0 E^\dagger$. Furthermore, this transformation also removes the highly oscillating part of $\hat{\xi}$,

($|\hat{\mu}_{lk}-\mu_{lk}| \ll \mu_{mn}$ and $\Gamma \ll A_{lk}\mu_{lk}$ for all l,k,m,n):

$$\frac{d}{dt}\hat{\zeta} = -i\sum_{lk}\frac{A_{lk}(\hat{\mu}_{lk}-\mu_{lk})}{2}\left[U^\dagger E\sigma_x^{lk}E^\dagger U,\hat{\zeta}\right] + \Gamma\sum_{j=1}^{N}\mathrm{Tr}\left(U^\dagger E P_j E^\dagger U(\zeta-\hat{\zeta})\right)\times \left(U^\dagger E P_j E^\dagger U\hat{\zeta}+\hat{\zeta}U^\dagger E P_j E^\dagger U - 2\mathrm{Tr}\left(U^\dagger E P_j E^\dagger U\hat{\zeta}\right)\hat{\zeta}\right).$$

Now let us develop the terms in the first part of (33), using this unitary transformation:

$$\mathrm{Tr}\left(\sigma_z^{lk}(\xi-\hat{\xi})\right)\mathrm{Tr}\left(\sigma_y^{lk}(\xi-\hat{\xi})\right) = \mathrm{Tr}\left(U^\dagger E\sigma_z^{lk}E^\dagger U(\zeta-\hat{\zeta})\right)\mathrm{Tr}\left(U^\dagger E\sigma_y^{lk}E^\dagger U(\zeta-\hat{\zeta})\right) = i\Big(\sum_{r\neq s}\exp(i(\Omega_r-\Omega_s)t)(E_{rl}E_{sl}-E_{rk}E_{sk})(\zeta_{sr}-\hat{\zeta}_{sr}) + \sum_r (E_{rl}^2-E_{rk}^2)(\zeta_{rr}-\hat{\zeta}_{rr})\Big)\times \left(\sum_{r\neq s}\exp(i(\Omega_r-\Omega_s)t)(E_{rl}E_{sk}-E_{rk}E_{sl})(\zeta_{sr}-\hat{\zeta}_{sr})\right).$$

Developing and removing the highly oscillating terms of frequencies Δ_Ω, we find

$$\sum_{r\neq s}(E_{rl}E_{sl}-E_{rk}E_{sk})(E_{rk}E_{sl}-E_{rl}E_{sk})|\zeta_{sr}-\hat{\zeta}_{sr}|^2 = \frac{1}{2}\sum_{r\neq s}((E_{rl}E_{sl}-E_{rk}E_{sk})(E_{rl}E_{sk}-E_{rk}E_{sl})|\zeta_{rs}-\hat{\zeta}_{rs}|^2 + (E_{rl}E_{sl}-E_{rk}E_{sk})(E_{sl}E_{rk}-E_{sk}E_{rl})|\zeta_{sr}-\hat{\zeta}_{sr})^2| = 0,$$

where we have broken the sum into two parts by symmetrizing with respect to the indices r and s.

Even though this argument does not prove the convergence of the estimator for the multi-level system, it gives a strong reason for it to be efficient.

6 Conclusion

In this paper, we propose an observer-based method for the Hamiltonian identification of a quantum system. A symmetry-preserving observer (6)-(7)-(8) has been developed to give an estimate of the unknown parameters of the 2-level system. Applying the averaging arguments, a complete but local analysis of the convergence for the estimation algorithm has been given. Since the observer design is based on the physical symmetries of the system, a multi-level extension of the estimator is straightforward. The convergence of this extension has been formally discussed. Various simulations in two or three dimensions illustrate the relevance and the robustness of the technique. Similar simulations can be performed for higher dimension systems.

Here let us finish the paper with some concluding remarks. Note that the non-degeneracy assumption for the Rabi transitions Δ_Ω in Section 5 may be removed using a slow modulation of the amplitudes A_{lk}. Furthermore, one does not really need to have access to the continuous measurement results $y_j(t)$ (which is lots of information to be asked in the laboratory settings). In fact, one only needs samples on the output signal with frequencies much higher than the larger Rabi frequency.

A Symmetry-preserving observers

In this section we recall the basic definitions and results of [5]. Consider the smooth system

$$\frac{d}{dt}x = f(x,u) \qquad \text{(A.1)}$$
$$y = h(x,u) \qquad \text{(A.2)}$$

where x belongs to an open subset $\mathscr{X}\subset\mathbb{R}^n$, u to an open subset $\mathscr{U}\subset\mathbb{R}^m$ and y to an open subset $\mathscr{Y}\subset\mathbb{R}^p$, $p\leq n$. We assume the signals $u(t),y(t)$ known. In section 3.2 we took

$$x=\rho,\quad u=(\sigma_x,\sigma_y,\sigma_z),\quad y=\mathrm{Tr}(\sigma_z\rho)$$

Consider also the local group of transformations on $\mathscr{X}\times\mathscr{U}$ defined by

$$(X,U) = \left(\varphi_g(x),\psi_g(u)\right), \qquad \text{(A.3)}$$

where φ_g and ψ_g are local diffeomorphisms depending on a parameter g which is an element of a Lie group G such that

- $\varphi_e(\xi)=\xi$ for all $\xi\in\mathscr{X}$
- $\varphi_{g_2}\left(\phi_{g_1}(\xi)\right)=\varphi_{g_2g_1}(\xi)$ for all $g_1,g_2\in G,\xi\in\mathscr{X}$.

and ψ_g verifies similar conditions. The action is said to be trivial if φ_g is the identity function. In section 3.2 we have $G=SU(2)$ and for any $U\in G$

$$\varphi_U=\psi_U:\ M\in\mathbb{C}^{2\times2}\mapsto UMU^\dagger$$

Definition 2 *The system $\frac{d}{dt}x=f(x,u)$ is G-*invariant *if $f\left(\varphi_g(x),\psi_g(u)\right)=D\varphi_g(x)\cdot f(x,u)$ for all g,x,u.*

The property also reads $\frac{d}{dt}X=f(X,U)$, i.e., the system remains unchanged under the transformation (A.3).

Definition 3 *A vector field w on $\mathscr{X}$ is said to be G-invariant if the system $\frac{d}{dt}x=w(x)$ is invariant. This means $w(\varphi_g(x))=D\varphi_g(x)\cdot w(x)$ for all g, x.*

Definition 4 *An invariant frame* $(w_1,\ldots,w_n)$ *on* $\mathscr{X}$ *is a set of n linearly point-wise independent G-invariant vector fields, i.e* $(w_1(x),\ldots,w_n(x))$ *is a basis of the tangent space to* $\mathscr{X}$ *at x.*

Instead of using the usual linear output error $\hat{y}-y$ we use an invariant output error:

Definition 5 *The smooth map* $(\hat{x},u,y)\mapsto E(\hat{x},u,y)\in\mathbb{R}^p$ *is an* invariant output error *if*

- *the map* $y\mapsto E(\hat{x},u,y)$ *is invertible for all* $\hat{x},u$
- $E\big(\hat{x},u,h(\hat{x},u)\big)=0$ *for all* $\hat{x},u$
- $E\big(\varphi_g(\hat{x}),\psi_g(u),h(\varphi_g(x),\psi_g(u))\big)=E(\hat{x},u,y)$ *for all* $\hat{x},u,y$

Definition 6 (pre-observer) *The system* $\frac{d}{dt}\hat{x}=F(\hat{x},u,y)$ *is a* pre-observer *of* (A.1)-(A.2) *if for all* x,u $F\big(x,u,h(x,u)\big)=f(x,u)$.

The definition does not deal with convergence; if moreover $\hat{x}(t)\to x(t)$ as $t\to+\infty$ for every (close) initial conditions, the pre-observer is an (asymptotic) *observer*.

Definition 7 *The pre-observer* $\frac{d}{dt}\hat{x}=F(\hat{x},u,y)$ *is* G-invariant *if for all* $g,\hat{x},u,y$,

$$F\big(\varphi_g(\hat{x}),\psi_g(u),h(\varphi_g(x),\psi_g(u))\big)=D\varphi_g(\hat{x})\cdot F(\hat{x},u,y).$$

The property also reads $\frac{d}{dt}\hat{X}=F(\hat{X},U,h(X,U))$, with $X=\varphi_g(x)$, $U=\psi_g(u)$. Assume that the output map is G-equivariant, which is the case for the quantum mechanical system considered in this paper since the output y is a scalar invariant. Then a sufficient condition for the system $\frac{d}{dt}\hat{x}=F(\hat{x},u,y)$ to be a G-invariant pre-observer for the G-invariant system $\frac{d}{dt}x=f(x,u)$ is:

$$F(\hat{x},u,y)=f(\hat{x},u)+\sum_{i=1}^{n}\mathscr{L}_i\big(I(\hat{x},u),E(\hat{x},u,y)\big)w_i(\hat{x}) \quad \text{(A.4)}$$

where E is an invariant output error, $(\hat{x},u)\mapsto I(\hat{x},u)\in\mathbb{R}^{n+m-r}$ is a full-rank invariant function, the $\mathscr{L}_i$'s are smooth functions such that for all $\hat{x}$, $\mathscr{L}_i\big(I(\hat{x},u),0\big)=0$, and $(w_1,\ldots,w_n)$ is an invariant frame. This result is a consequence of the theorem 2 of [5]. I is called a complete set of scalar invariants and verifies $I(\varphi_g(\hat{x}),\psi_g(u))=I(\hat{x},u)$ for any $g\in G$.

Thus, to build a symmetry-preserving observer, one needs a) an invariant output error b) an invariant frame c) a complete set of scalar invariants.

B Identifiability

In this appendix, we present the mathematical framework in which the identification problem can be considered. Moreover, we review briefly the former work on the identifiability of the considered system. A well-posedness result which allows us to consider the identification problem in Section 5 will be announced.

The goal is to identify $H=H_0+V$ or/and μ in system

$$i\frac{d}{dt}\Psi=(H_0+V+u(t)\mu)\Psi,\quad \Psi|_{t=0}=\Psi_0,\quad \|\Psi_0\|_{\mathscr{H}}=1, \tag{B.1}$$

when laboratory measurements on some physical observables are provided. In [21], two different settings have been considered in order to characterize the identifiability of such a system:

(S1) The Hamiltonian H is known and the goal is to identify the dipole moment μ. The so-called *populations* along the eigenstates ϕ_i, i.e. $p_i=|\langle\phi_i,\Psi(t)\rangle|^2, i=1,2,\ldots,N$ are measured for all instants $t\geq 0$. This is performed with as many control amplitudes $u(t)$ as required.

(S2) Neither the potential V nor the dipole moment μ are known and the goal is to identify them. Note that, by identifying H we mean identifying V, as H_0 is readily known. The eigenvalues of the Hamiltonian $H=H_0+V$ are also assumed to be known (this assumption is relevant in practice, see Remark 8). Here we measure the populations p_i along the states of a canonical basis $\{e_i\}_{i=1}^N$: $p_i=|\langle e_i,\Psi(t)\rangle|^2, i=1,2,\ldots,N$ for all instants $t>0$ and all control amplitudes $u(t)$.

Remark 8 *It is relevant in practice to assume that the eigenvalues of the internal Hamiltonian* $H=H_0+V$ *are known. In fact the classical spectroscopy allows for identifying the eigenvalues of the Hamiltonian and discriminating between two systems that do not share the same ones. In fact spectroscopy only gives eigenvalue differences (transition frequencies), not the absolute values. The overall unknown additive factor is not seen by the measurements and has no impact on the identification result.*

In this paper, we have only considered the first setting. An extension of the technique to the second setting remains to be done in future work. However, [21] provides an identifiability result for this second setting as well.

Here we announce the identifiability result of [21] concerning the first setting. For a result in the second setting and also the proof of the result for the first setting, we refer to [21].

Theorem 9 *Suppose that there exist two dipole moments* μ_1 *and* μ_2*, giving rise to two evolving states* Ψ_1 *and* Ψ_2 *respectively solving*

$$i\frac{d}{dt}\Psi_1=(H+u(t)\mu_1)\Psi_1, \tag{B.2}$$

$$i\frac{d}{dt}\Psi_2=(H+u(t)\mu_2)\Psi_2, \tag{B.3}$$

that produce identical observations for all $t\geq 0$ *and all fields*

$u(t)$:

$$|\langle\Psi_1(t),\phi_i\rangle|^2 = |\langle\Psi_2(t),\phi_i\rangle|^2 \qquad i=1,2,...,N. \qquad \text{(B.4)}$$

Then under assumptions

(A1) *Equation* (B.2) *is wave-function controllable [34];*
(A2) *The transitions of the Hamiltonian H are non-degenerate: $\lambda_{i_1}-\lambda_{j_1}\neq\lambda_{i_2}-\lambda_{j_2}$ for $(i_1,j_1)\neq(i_2,j_2)$ [38];*
(A3) *The diagonal part of the dipole moments μ_1 and μ_2, when written in the eigenbasis of the Hamiltonian H, is zero: $\langle\phi|_i\,\mu_1\,|\phi\rangle_i = \langle\phi|_i\,\mu_2\,|\phi\rangle_i = 0, i=1,2,...,N$;*

the two dipole moments are equal within some phase factors $\{\alpha_i\}_{i=1}^N\subset\mathbb{R}$ such that:

$$\forall i,j=1,2,...,N, \qquad (\mu_1)_{ij} = e^{i(\alpha_i-\alpha_j)}(\mu_2)_{ij}. \qquad \text{(B.5)}$$

Fore more details and remarks concerning the assumptions and the result of this theorem we refer to [21].

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