

Quantum state reconstruction for rigid rotors

Sarin A. Deshpande, Gregory S. Ezra *

Department of Chemistry and Chemical Biology, Baker Laboratory, Cornell University, Ithaca, NY 14853, United States

Received 26 December 2006; in final form 23 March 2007

Available online 19 April 2007

Abstract

We describe a quantum state reconstruction scheme for dipolar rigid rotors based on determination of the expectation value of the molecular orientation. A key feature is the use of half-cycle pulses to excite the rotor prior to the orientation measurement. The set of expectation values obtained by varying the intensity and polarization of the laser and the time interval between excitation and measurement can be inverted directly to yield the rotor density operator. When the density operator corresponds to the admixture of relatively few rotor states, our procedure successfully reconstructs both pure and mixed states.

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1. Introduction

The general question of inferring or reconstructing the quantum state of a system from measurement of expectation values of observables was raised early on by Pauli [1–4]. There has been renewed recent interest in the problem, particularly in the development of quantum tomography (see, for example, Refs. [5–7]).

In this Letter we consider the problem of reconstruction of the quantum state (density operator) of a dipolar rigid rotor molecule using as input the expectation values of relevant observables. Any density operator of finite rank N can be written as a linear combination of N^2 Hermitian operators that form a basis in an operator inner-product space [8]. The expectation values of the N^2 basis operators then determine the density operator uniquely. From the practical point of view, the problem is that the relevant basis operators will in general correspond to complicated observables whose physical measurement may be difficult to perform. In the case of a rotor, for example, the operator basis will consist of irreducible spherical tensor opera-

tors of high rank [9,10]. A previously suggested approach for reconstruction of rotor states implicitly requires the measurement of tensor operators of high rank [11] (see also Ref. [12]).

Here we describe a reconstruction scheme for the rigid rotor that relies solely on determination of the expectation value of a low rank tensor, in this case the molecular orientation. Previous work on the determination of spin quantum states has shown that a set of $4F + 1$ Stern–Gerlach measurements with respect to suitably chosen quantization axes serve to define the quantum state of a spin F [13,2,14] (see also [15]). A key feature of our approach is the use of one or more ultrashort half-cycle pulses (HCPs) [16–20] to excite the rotor prior to the time at which the measurement is made. The action of the pulse on the rotor is assumed known; we shall use the sudden approximation for simplicity [21,22]. By varying the intensity and polarization of the laser, and the time interval between excitation and measurement, we obtain a set of expectation values that can be inverted directly to yield the rotor density operator. In the case that the density operator corresponds to the admixture of relatively few rotor states, our procedure is shown to successfully reconstruct both pure and mixed density operators. Possible extensions of our approach to more complicated cases are discussed in the Summary section.

* Corresponding author. Fax: +1 607 255 4137.

E-mail address: gse1@cornell.edu (G.S. Ezra).

2. Quantum state reconstruction via matrix inversion

2.1. General principles

A general state of an N -state quantum system is described by the $N \times N$ density matrix ρ_{ij} , $i, j = 1, \dots, N$, where, if ρ is normalized ($\text{tr}[\rho] = 1$), a set of $(N^2 - 1)$ real parameters characterize the matrix ρ [8]. The density operator ρ can in general be expressed as a linear combination of $(N^2 - 1)$ operators that form a basis in a space of operators [8], and the coefficients in the expansion can then be determined by determining expectation values (traces) of the $N^2 - 1$ operators in the state ρ [8].

For a problem involving rotor states a natural operator basis consists of irreducible spherical tensor operators constructed by coupling the ket and bra sides of the density operator [9,10]. Complete determination of the density operator then in general requires the evaluation of expectation values of tensor operators of large rank. Essentially such an approach has been proposed by Mouritzen and Molmer, who have shown that the density operator of a rigid rotor can in principle be determined by measurement of the orientational distribution function of the rotor axis [11]. Resolution of fine details of the angular distribution then corresponds to measurement of the expectation values of high-order tensor operators.

It is natural to ask whether it is possible to determine the density operator for a rigid rotor using a set of measurements of simple observables (i.e., tensor operators of low rank), such as orientation (expectation value of $\cos\theta$, where θ is the angle between the rotor dipole axis and the lab-fixed z -direction). The measurements are to be made on an ensemble of identically prepared systems described by the density operator ρ .

Suppose the system, initially in state ρ , is subjected to an ultrashort pulse at $t = 0$, which transforms the density matrix essentially instantaneously from $\rho \rightarrow \bar{\rho}$. For an impulsive perturbation of intensity I_p , the transformed density operator is

$$\bar{\rho}(p) = \hat{V}(p)\rho\hat{V}(p)^\dagger, \quad (1a)$$

or, in some orthonormal basis

$$\bar{\rho}(p)_{ij} = \sum_{i'j'} V(p)_{ii'} \rho_{i'j'} V(p)_{jj'}^\dagger, \quad (1b)$$

where the transformation matrix $V(p)_{ij}$ is assumed known (see below). The state at time $t = t_n$ after the pulse is then

$$\bar{\rho}(p, n) = \hat{U}_0(t_n)\hat{V}(p)\rho\hat{V}(p)^\dagger\hat{U}_0(t_n)^\dagger \quad (2)$$

where $\hat{U}_0(t_n)$ is the operator representing free propagation for time t_n . If observable $\hat{\Omega}$ is measured at time t_n , the expectation value $\langle\Omega\rangle(k) \equiv \langle\Omega\rangle(p, n)$ is

$$\langle\Omega\rangle(k) \equiv \langle\Omega\rangle(p, n) = \text{tr}[\hat{\Omega}\hat{U}_0(t_n)\hat{V}(p)\rho\hat{V}(p)^\dagger\hat{U}_0(t_n)^\dagger] \quad (3a)$$

$$= \sum_{ij} (\hat{V}(p)^\dagger\hat{U}_0(t_n)^\dagger\hat{\Omega}\hat{U}_0(t_n)\hat{V}(p))_{ij}\rho_{ji} \quad (3b)$$

$$\equiv \sum_{ij} M(k)_{ij}\tilde{\rho}_{ij} \quad (3c)$$

$$\equiv \mathbf{M}(k) \cdot \tilde{\boldsymbol{\rho}} \quad (3d)$$

where in the last line we have written the expectation value $\langle\Omega\rangle(k)$ as the inner-product of the N^2 -dimensional vectors $\mathbf{M}(k)$ and $\tilde{\boldsymbol{\rho}}$. If we assume that the density operator is effectively a finite-dimensional $N \times N$ matrix, then by taking various combinations of intensity I_k and post-pulse propagation times t_n , we can in principle obtain a set of linear equations for the initial state density matrix elements ρ_{ij} , which can be solved to determine ρ . If ρ is effectively an $N \times N$ matrix, then taking N^2 combinations (rather than $N^2 - 1$, as we shall not explicitly impose a normalization condition) of pulse intensity I_p and observation time t_n we obtain an $N^2 \times N^2$ measurement supermatrix

$$\mathcal{M}_{k,\ell} \equiv M(k)_{ij}, \quad (4)$$

where $k \equiv (p, n) = 1, \dots, N^2$, $\ell \equiv (ij) = 1, \dots, N^2$. The matrix \mathcal{M} is independent of the initial state ρ ; if \mathcal{M} is invertible (full rank), then we can solve the set of N^2 linear equations

$$\langle\Omega\rangle(k) = \sum_{\ell=1}^{N^2} \mathcal{M}_{k,\ell}\tilde{\rho}_\ell \quad k = 1, \dots, N^2 \quad (5)$$

directly to determine the matrix elements ρ_{ij} . Note that in the practical implementation of the theory it is not necessary to impose any constraints on the matrix elements ρ_{ij} ; both normalization and pure/mixed character of the density operator are completely determined by the algorithm.

We demonstrate below that it is possible to obtain invertible measurement supermatrices for dipolar rigid rotors subject to HCP followed by measurement of orientation. In that case, we can invert a set of Eq. 5 to reconstruct the quantum state ρ , provided the effective size of ρ is not too large.

2.2. Quantum state reconstruction for dipolar rigid rotors

In our approach we imagine the rigid rotor to be excited using an ultrashort HCP at $t = 0$ [17–20], and expectation values of the orientation $\hat{\Omega}$ of the molecule evaluated at various times. Recent experiments have shown the feasibility of measuring the orientation $\langle\cos\theta\rangle$ of dipolar molecules like OCS on femtosecond timescales [23,24]. In general we need to consider a set of expectation values obtained using different combinations of laser polarization vector $\hat{\mathbf{n}}$, laser intensity I_p , and measurement time t_n .

We consider excitation pulses for which the pulse duration T_p is much shorter than the rotational period T_{rot} [17–20], so that the interaction can be treated using the sudden approximation. This approximation is not essential, but provides an analytical model for the rotor excitation dynamics [21]. If the sudden approximation is not applicable, it is necessary to integrate the quantum mechanical equations of motion for the time-dependent Hamiltonian

Eq. (6) over the duration of the pulse. The molecule-laser Hamiltonian is

$$\hat{H}(t) = \hat{H}_0 + \hat{H}'(t) \quad (6)$$

with free rotor Hamiltonian

$$\hat{H}_0 = B\hat{J}^2, \quad (7a)$$

(rotational constant B) and molecule-laser interaction

$$\hat{H}'(t) = -E_0 \mathbf{d} \cdot \hat{\mathbf{n}}g(t) \quad (7b)$$

where $\hat{\mathbf{n}}$ is the laser polarization vector, \mathbf{d} is the rotor dipole operator, and the magnitude of the laser electric field is $E(t) = E_0g(t)$. We take the pulse profile $g(t)$ to be a cosine lobe of width $T_p/2$, where $T_p \approx 1$ ps is the period of the full pulse cycle, corresponding to a frequency $\nu_p = 1$ THz [16]. In the sudden approximation, matrix elements between free-rotor states $|jm\rangle$ and $|j'm'\rangle$ of the complete propagator over the duration of the pulse are approximated as follows [21]:

$$\begin{aligned} & \left\langle j'm' \left| \exp \left[-\frac{i}{\hbar} \int_{-T_p/4}^{+T_p/4} dt \hat{H}(t) \right] \right| jm \right\rangle \\ & \simeq \left\langle j'm' \left| \exp \left[-\frac{i}{\hbar} \int_{-T_p/4}^{+T_p/4} dt \hat{H}'(t) \right] \right| jm \right\rangle \end{aligned} \quad (8a)$$

$$= \left\langle j'm' \left| \exp \left[+i\beta \hat{\mathbf{d}} \cdot \hat{\mathbf{n}} \right] \right| jm \right\rangle \quad (8b)$$

where the parameter β is

$$\beta = \frac{dE_0}{\hbar} \int_{-T_p/4}^{+T_p/4} dt g(t) = \frac{dE_0}{\hbar} \int_{-T_p/4}^{+T_p/4} dt \cos 2\pi\nu_p t = \frac{2dE_0}{h\nu_p} \quad (9)$$

and $\hat{\mathbf{d}}$ is a unit vector along the molecular dipole axis.

The matrix V that transforms ρ to $\bar{\rho}$ in the free-rotor basis is then

$$\langle j'm' | V | jm \rangle = \langle j'm' | \exp[+i\beta \hat{\mathbf{d}} \cdot \hat{\mathbf{n}}] | jm \rangle \quad (10a)$$

$$= \langle j'm' | \hat{R}(\phi_n, \theta_n, 0) \exp[+i\beta \hat{\mathbf{d}} \cdot \hat{\mathbf{z}}] \hat{R}^\dagger(\phi_n, \theta_n, 0) | jm \rangle \quad (10b)$$

$$= \langle j'm' | \hat{R}(\phi_n, \theta_n, 0) \exp[+i\beta \cos \theta] \hat{R}^\dagger(\phi_n, \theta_n, 0) | jm \rangle \quad (10c)$$

where \hat{R} is the unitary rotation operator that rotates the lab-fixed z -axis into the laser polarization axis $\hat{\mathbf{n}}$ [10]. The rotation operator depends on only two Euler angles (θ_n, ϕ_n) , the spherical polar angles defining the direction of the polarization axis $\hat{\mathbf{n}}$.

If the rotor density operator is effectively an $N \times N$ matrix ρ , N^2 measurements must be made to define the $N^2 \times N^2$ measurement supermatrix \mathcal{M} . For a tractable reconstruction problem, the effective size of the state space N should not be too large. Moreover, for theoretical analysis the pulse at $t = 0$ should not excite too large a number of excited rotor states, as we wish to ensure that the quantum mechanical calculation of the excitation process and subsequent time evolution is converged. The pulse should not be too weak, however, as this can lead to numerical difficulties inverting \mathcal{M} to obtain ρ (see below).

For experimentally available HCP, $T_p/2 \approx 500$ fs [16], so that for most molecules (except hydrides such as HF and HCl) the condition $T_p/2 \ll T_{\text{rot}}$ is satisfied, and the sudden approximation provides a reasonably accurate description of the excitation process. Attainable maximum field strengths are of order $E_0 \sim 1.5 \times 10^7$ V/m [16], so that for a molecule with a large dipole moment, such as LiCl ($d = 7.13$ D [25]), the corresponding value of β is $\beta \approx 1.5$. In the calculations presented below we implement our scheme for laser intensities corresponding to β values in the range $0 < \beta < \beta_{\text{max}} = 1.5$. Reducing the value of β_{max} leads to a larger condition number for \mathcal{M} , and possibly less accurate reconstruction of the density matrix (see below). For molecules having appreciably smaller dipole moments than LiCl, either larger HCP peak field strengths E_0 must be used to obtain the same value of β_{max} , or the scheme can be extended to allow for a sequence of HCPs acting on the molecule before measurement of orientation (see Section 4).

3. Theoretical implementation of reconstruction scheme

3.1. Mixture of j states at constant $m = 0$

Consider the case of a mixture of rotor states with fixed m , $m = 0$ say. For a z -polarized laser field, we have the relatively simple expression for the matrix elements (10) [21]

$$\begin{aligned} V_{j'j} & \equiv \langle j'0 | V | j0 \rangle \\ & = \sqrt{(j+1)(j'+1)} \sum_l i^l j_l(\beta) \langle j'0j'0 | l0 \rangle^2, \end{aligned} \quad (11)$$

where $\langle j_1, m_1, j_2, m_2 | j_3, m_3 \rangle$ are Clebsch–Gordan (vector coupling) coefficients [10] and $j_l(\beta)$ are spherical Bessel functions, with intensity parameter β being related to the laser field strength by Eq. (9).

To illustrate our reconstruction method, an initial density operator ρ is obtained as follows. A rotor in the ground state $\rho_0 = |0,0\rangle\langle 0,0|$ is subject to a z -polarized HCP with $\beta = 1.0$. This excitation process defines a pure initial state $\rho = \hat{V} \rho_0 \hat{V}^\dagger$ with $m = 0$ for subsequent analysis. To obtain a set of mixed density operators for analysis, the off-diagonal elements of ρ in the free-rotor basis are multiplied by an attenuation factor γ^ν , where $\gamma = \frac{1}{\sqrt{2}}$ and $\nu = 1, \dots, 4$. The set of five density operators $\rho^{(\nu)}$, $\nu = 0, 1, \dots, 4$ is then used to compute ‘experimental’ $\langle \cos \theta \rangle$ values for a combination of different times (t_n) and intensities $I_p(\beta)$, ($0 \leq \beta \leq \beta_{\text{max}} = 1.5$), and the quantum state determination scheme applied to this numerically generated data for each of the initial states $\rho^{(\nu)}$.

The size of the measurement supermatrix \mathcal{M} is determined by the maximum assumed value of the rotor quantum number, j_{max} . For each value of $j_{\text{max}} \equiv N' - 1$ we obtain an $N'^2 \times N'^2$ matrix $\mathcal{M}(N')$ by taking suitable combinations of intensity and measurement times; if the matrix $\mathcal{M}(N')$ can be inverted, we obtain the (transpose of) the reconstructed density matrix

$$\tilde{\rho}_{\text{rec}}(N') = \mathcal{M}(N')^{-1} \cdot \mathbf{\Omega}(N'). \quad (12)$$

where $\mathbf{\Omega}(N')$ represents the N'^2 -dimensional vector of measured orientations, $\langle \cos \theta \rangle$.

We have explored numerically several excitation/measurement schemes. For example, matrices \mathcal{M} were obtained using N'^2 values of t_n for fixed intensity parameter β . Also, calculations have been carried out with the observation time t_n fixed and N'^2 different β values. In both cases it is found that accurate inversion of the measurement matrix is only possible using unphysically large values of β_{max} . These excitation/measurement schemes are however found to work for physically more reasonable values of β when more than one excitation pulse is used (cf. Section 4).

For the calculations reported here, we used a set of N'^2 randomly chosen pairs of values $\{t_k, \beta_k\}$ to obtain the observation matrix \mathcal{M} , taking observation times in the range $0 \leq t_n \leq t_{\text{max}} = T_{\text{rot}}$, and laser intensities such that $0 \leq \beta \leq \beta_{\text{max}} = 1.5$.

The convergence of the reconstructed density operator $\rho_{\text{rec}}(N')$ to the original ρ can be studied in several ways. In Fig. 1 we plot the trace $\text{tr}[\rho_{\text{rec}}(N')]$ as a function of N' ; this plot indicates that the reconstruction is essentially converged at $j_{\text{max}} = 4$. (It should be noted that convergence of all calculations is also checked with respect to the *total*

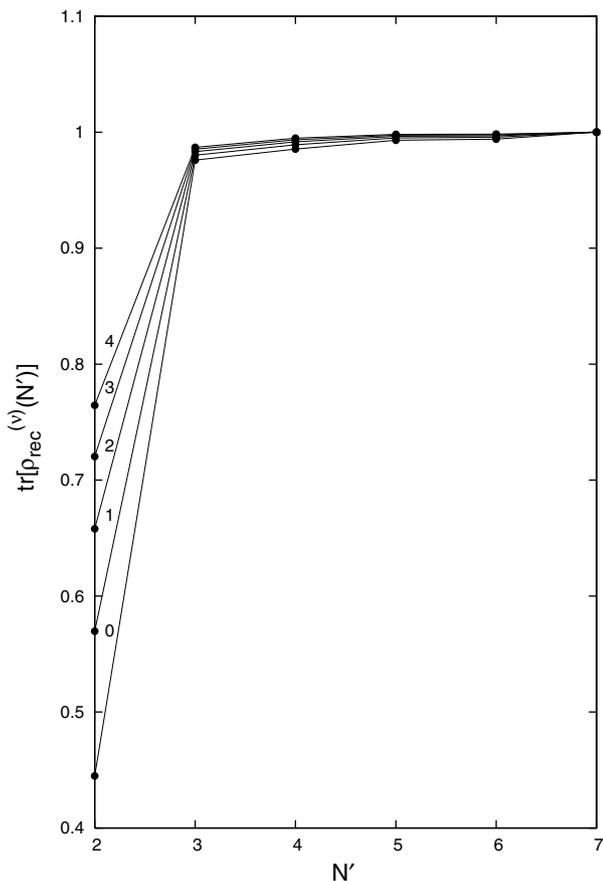


Fig. 1. Trace $\text{tr}[\rho_{\text{rec}}^{(v)}(N')]$ of the reconstructed density operator $\rho_{\text{rec}}^{(v)}(N')$ as a function of the assumed density operator rank N' , for $v=0-4$. The maximum intensity parameter $\beta_{\text{max}} = 1.5$. This plot indicates that the reconstruction is essentially converged at $j_{\text{max}} = N' - 1 = 4$.

rotor basis size N_{tot} ; for the present example, use of a basis of rotor states with $N_{\text{tot}} = 13$ ensures that all expectation values are converged with respect to basis size.) Fig. 2 shows $\text{tr}[\rho_{\text{rec}}(N')^2]$ as a function of N' . For the mixed states, $\text{tr}[\rho_{\text{rec}}^2] < 1$. Again we see convergence by $j_{\text{max}} = 4$. Finally, Fig. 3 shows convergence of the norm of the difference

$$\|\rho - \rho_{\text{rec}}(N')\|^2 \equiv \sum_{i,j=1}^{N'} |(\rho - \rho_{\text{rec}}(N'))_{ij}|^2 \quad (13)$$

with increasing N' .

We now consider in more detail the dependence of the accuracy of the reconstruction procedure on the maximum intensity parameter β_{max} for $j_{\text{max}} = 6$. In Fig. 4a we show the condition number of the measurement matrix \mathcal{M} (defined as the square root of the absolute magnitude of the ratio of the largest to the smallest eigenvalue of $\mathcal{M}^\dagger \mathcal{M} = \widetilde{\mathcal{M}}^* \mathcal{M}$) as a function of β_{max} . Although the matrix \mathcal{M} is quite poorly conditioned even for $\beta_{\text{max}} = 1.5$, straightforward numerical inversion succeeds in reproducing the density operator to acceptable accuracy for β_{max} values down to ≈ 0.05 . This is seen in Fig. 4b, which plots $\log(\|\rho - \rho_{\text{rec}}\|^2)$ versus $\log \beta_{\text{max}}$. For smaller values of β_{max} , use of the pseudoinverse [26] yields a reasonably accurate reconstructed density matrix even for the poorly condi-

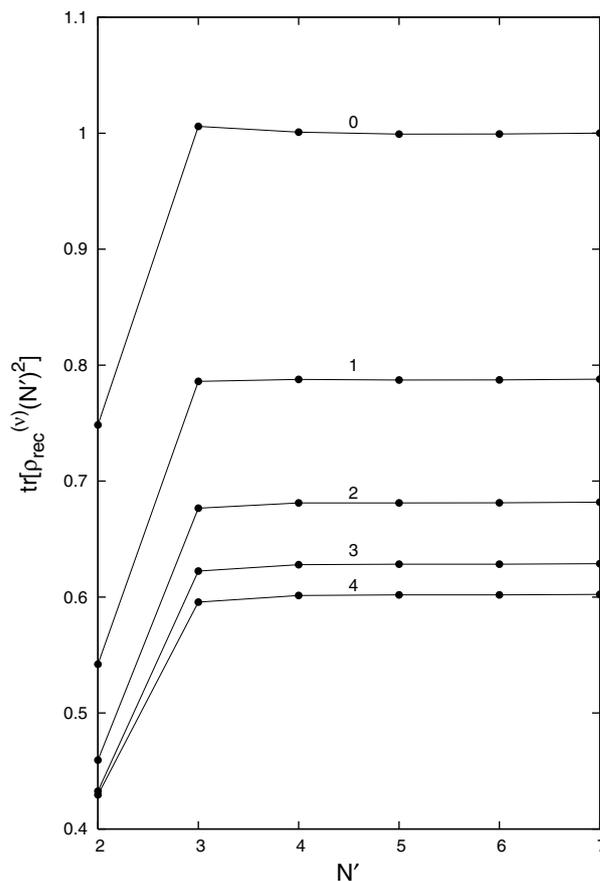


Fig. 2. Trace $\text{tr}[\rho_{\text{rec}}^{(v)}(N')^2]$ of the square of the reconstructed density matrix $\rho_{\text{rec}}^{(v)}(N')$ as a function of the assumed density operator rank N' , for $v=0-4$. The maximum intensity parameter $\beta_{\text{max}} = 1.5$. For mixed states, $v \geq 1$, $\text{tr}[\rho_{\text{rec}}^{(v)2}] < 1$. Again we see convergence by $j_{\text{max}} = 4$.

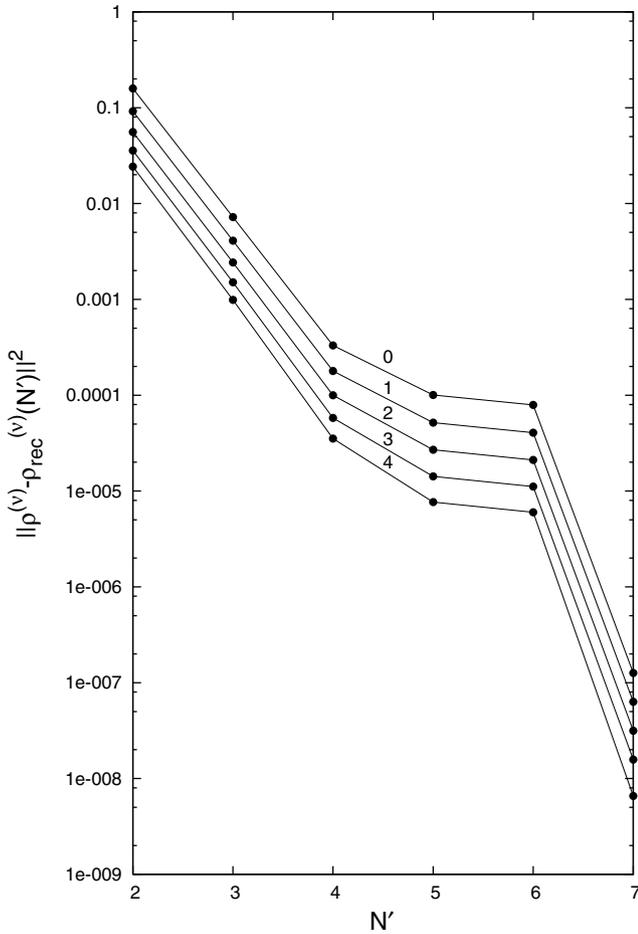


Fig. 3. Norm squared $\|\rho^{(v)} - \rho_{\text{rec}}^{(v)}(N')\|^2$ (cf. Eq. (13)) of the difference between the density operator $\rho^{(v)}$ and the reconstructed operator $\rho_{\text{rec}}^{(v)}(N')$ as a function of the assumed density operator rank N' , for $v=0-4$. Note the log scale. The maximum intensity parameter $\beta_{\text{max}} = 1.5$.

tioned measurement matrices obtained at β_{max} values as low as 0.03. (For use of the pseudoinverse in density matrix reconstruction, see also Ref. [14].)

It is shown in Section 4 that use of a multi-pulse excitation scheme serves to stabilize the inversion procedure.

3.2. Mixture of m states at constant j

For states that are mixtures of rotor states with fixed j , possible m values are $m = +j, j-1, \dots, -j$, so that the dimension of the state space is fixed in this case, $N = 2j + 1$. The measurement supermatrix \mathcal{M} is then a $(2j + 1)^2 \times (2j + 1)^2$ matrix.

Expressions for the matrix elements V (10a) are more complicated than in the previous case with $m = 0$. We have

$$V_{j_1 m_1, j m} \equiv \langle j_1 m_1 | V | j m \rangle \quad (14a)$$

$$\begin{aligned} &= \sum_{\bar{m}} (-1)^{-\bar{m}} e^{i\phi_n(m-m_1)} d_{m_1 \bar{m}}^{j_1} \left(\frac{\theta_n}{2} \right) d_{\bar{m} m}^j \left(-\frac{\theta_n}{2} \right) \\ &\times \sum_l i^l j_l(\beta) \sqrt{(j_1 + 1)(j + 1)} \\ &\times \langle j_1 - \bar{m} j \bar{m} | 0 \rangle \langle j_1 0 j 0 | 0 \rangle \end{aligned} \quad (14b)$$

where we have used the fact that $\langle j_1 - \bar{m} j \bar{m} | 0 \rangle = \delta_{\bar{m} j \bar{m}}$.

The symmetry property of the rotation matrices [10]

$$d_{m_1 m}^j \left(\frac{\theta}{2} \right) = (-1)^{(m_1 - m)} d_{-m_1 -m}^j \left(\frac{\theta}{2} \right) \quad (15)$$

implies certain relations between elements of the \mathcal{M} matrix, which lead to difficulties in the implementation of the reconstruction method. For example, for any laser polarization vector \hat{n} and for $\hat{\Omega} = \cos \theta$ it can be shown that

$$\mathcal{M}_{j m, j m} = \mathcal{M}_{j -m, j -m}. \quad (16)$$

The measurement matrix \mathcal{M} therefore does not have full rank (it has two identical columns) and cannot be inverted to obtain ρ . In order to get around this problem we consider expectation values of two observables, $\hat{\Omega}_1 = \hat{z} = \cos \theta$ and $\hat{\Omega}_2 = \hat{x} = \sin \theta \cos \phi$, corresponding to measurement of the rotor orientation along the z - or the x -axis, respectively. If the observation set includes measurements of orientation along both the x - and z -axes, then an invertible matrix \mathcal{M} can be obtained.

As an example, consider the case $j = 2$. As before we construct a pure state density operator and obtain a set of mixed states by multiplying off-diagonal density matrix elements by an attenuation factor. The initial pure state is taken to be the rotor coherent state [27]

$$|j; \bar{\alpha} \bar{\beta}\rangle = \sum_{m=-j}^{+j} |j, m\rangle c_m = \sum_{m=-j}^{+j} |j, m\rangle D_{m j}^j(\bar{\alpha}, \bar{\beta}, 0) \quad (17)$$

specified by the Euler angles $(\bar{\alpha}, \bar{\beta}, 0)$. Matrix elements of the corresponding $(2j + 1) \times (2j + 1)$ density matrix are $\rho_{m m'} = c_m c_{m'}^*$. As previously, the pure state density matrix ρ was used to generate a set of mixed state density operators $\rho^{(v)}$ by multiplying off-diagonal matrix elements by the factor $(1/\sqrt{2})^v$, $v = 1, \dots, 5$.

Again, each of the matrices $\rho^{(v)}$ is used to obtain ‘experimental’ data for a combination of $(2j + 1)^2 = 25$ different laser polarization directions, intensities, observation times, and orientation axes. The calculations were performed on the density matrix derived from the coherent state with $\bar{\alpha} = \pi/\sqrt{2}$ and $\bar{\beta} = \pi/\sqrt{5}$. We chose to vary the time of observation t_n , the laser polarization angle θ_n (the angle ϕ_n is held fixed, $\phi_n = \pi/\sqrt{3}$), and the intensity. The reconstruction calculation was carried out in two different ways: first, 25 random values for time and polarization direction were chosen while keeping the intensity parameter β fixed, $\beta = 1.41$. Second, five observation times and five pairs of polarization angle and laser intensity were chosen, and 25 expectation values calculated for the direct product set of parameter values. Several such parameter sets were investigated, and a representative set of values is shown in Table 1. Using either approach the reconstructed density operator matches the original exactly. Results for the first method described above are given in Table 2. It can be seen that all the density matrices $\rho^{(v)}$ can be reconstructed essentially exactly by inverting Eq. 12 with $N' = 25$.

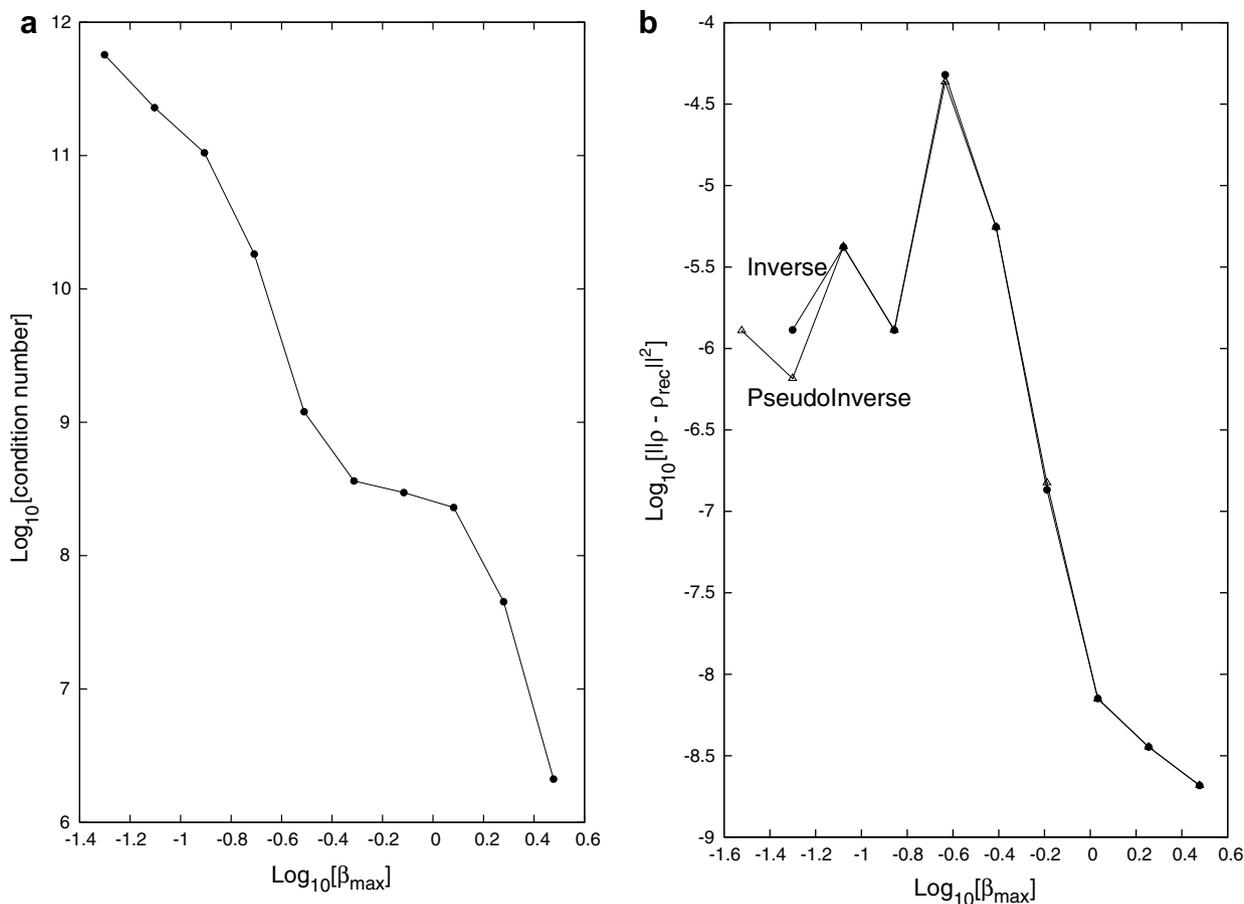


Fig. 4. (a) Log of the condition number of the measurement matrix $\mathcal{M}(\beta_{\max})$ as a function of the log of the intensity parameter β_{\max} . (b) $\log(\|\rho - \rho_{\text{rec}}\|^2)$ versus $\log \beta_{\max}$ obtained using either straightforward matrix inversion (filled circles) or the pseudoinverse (triangles).

Table 1

Values of time t_k , direction $\hat{n}(\theta_k, \phi)$ of the polarization laser, and laser intensity parameter β_k used in the inversion calculation for the case $j = 2$ discussed in Section 3.2

t_k	θ_k/π	β_k
0.5272	0.2276	0.2422
0.9133	0.7904	0.6371
0.6276	0.8441	1.3328
0.5752	0.1739	1.2456
0.0948	0.9745	1.2625

The angle ϕ is fixed at value $\phi = \pi/\sqrt{3}$.

Table 2

Properties of the reconstructed density operator $\rho_{\text{rec}}^{(v)}$ calculated for density operators based on the rotor coherent state Eq. (17) with $j = 2$, $v = 0-5$

v	$\text{tr}[\rho_{\text{rec}}^{(v)}]$	$\ \rho_{\text{rec}}^{(v)} - \rho^{(v)}\ ^2$	$\text{tr}[\rho_{\text{rec}}^{(v)2}]$
0	1.00	0.00	1.000
1	1.00	0.00	0.639
2	1.00	0.00	0.458
3	1.00	0.00	0.368
4	1.00	0.00	0.323
5	1.00	0.00	0.300

4. Multi-pulse excitation

As shown in the preceding section, although reconstruction of the density operator for rigid rotors is numerically feasible, the measurement supermatrix \mathcal{M} can be ill-conditioned, especially for small values of β_{\max} . This results in extreme sensitivity of the inversion procedure to random errors in the measurement of rotor orientation.

The first and simplest way to stabilize the inversion procedure is to increase the value of β_{\max} , corresponding physically to increasing the intensity of the exciting HCP. Another method is to excite the rotor with more than one pulse before making measurements of molecular orientation. In this case it is not necessary to increase the value of β_{\max} , so that β can be restricted to the range $0 \leq \beta \leq \beta_{\max} = 1.5$. Multi-pulse excitation corresponds to effectively larger values of β_{\max} , and leads to smaller condition numbers for the measurement matrix \mathcal{M} .

To illustrate, consider two-pulse excitation with $N' = 7$ and $N_{\text{tot}} = 13$. Calculations were performed by choosing $N^2 = 49$ triples $\{\beta_k, t_k^{(1)}, t_k^{(2)}\}$, where $0 \leq t_k^{(v)} \leq T_{\text{rot}}$ is the time at which pulse v interacts with the system, $v = 1, 2$, and $0 \leq \beta_k \leq \beta_{\max} = 1.5$. The $\text{Log}_{10}[\text{condition number}]$ for \mathcal{M} is reduced to ≈ 4 . With multi-pulse excitation, we

Table 3
Reconstruction in the presence of random observational errors for the two-pulse excitation scheme discussed in Section 4

A	$\text{tr}[\rho_{\text{rec}}]$	$\ \rho_{\text{rec}} - \rho\ ^2$	$\text{tr}[\rho_{\text{rec}}^2] - \text{tr}[\rho^2]$
10^{-6}	0.9999	2.62×10^{-7}	3.61×10^{-6}
10^{-5}	0.9999	2.59×10^{-5}	1.25×10^{-5}
10^{-4}	0.9991	2.59×10^{-3}	2.2×10^{-3}
10^{-3}	0.9913	0.2591	0.2553

A random Gaussian distributed error on the rotor orientation of magnitude A is added to the orientation expectation values.

are able to add random Gaussian distributed errors $\delta\Omega$ to the computed orientation values and still reconstruct the density matrix to a reasonable degree of accuracy (see Table 3).

5. Summary and conclusion

We have described and implemented a simple quantum state reconstruction scheme for rigid dipolar rotors. In contrast to other proposals for rotor state reconstruction [11], our method involves measurement of the expectation value of a low-rank tensor operator, the molecular orientation. Our approach requires a set of experiments to be performed in which the initial rotor state is excited by one or more ultrashort half-cycle laser pulses, followed by measurement of molecular orientation after a known time delay. Variation of the pulse intensities and polarization, the time delay and the axis along which the orientation is measured provides a set of measured values that are linear combinations of matrix elements of the rotor density matrix [8]. We have shown that it is possible to define a suitable set of measurements for which this relation can be inverted to yield the density matrix directly, at least in the case where ρ corresponds to the admixture of relatively few states. Further study of the invertibility of the measurement supermatrix \mathcal{M} for different excitation schemes would be desirable.

Our method is not necessarily restricted to rigid rotors, as rotor nonrigidity can be accounted for by including higher order centrifugal terms in the effective rotor Hamiltonian.

We now mention some limitations of our approach and possible avenues for future investigation. First, the possible extension of our scheme to nonlinear rotors (symmetric and asymmetric tops) and to nonpolar linear molecules and would be of interest. In present form our method is not practicable if a large number of levels are initially populated. (The number of density matrix elements to be deter-

mined scales as the square of the number of levels populated.) If many levels are populated, then it will be necessary to parametrize the density operator in some fashion. A maximum entropy approach [8] to determination of the density operator subject to constraints provided by observables of the kind discussed here is one possibility. The effects of decoherence and/or dissipation on the present scheme are also of great interest. Some explicit modeling of the effects of dephasing/dissipation will presumably be necessary, but this lies outside the scope of the present paper.

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