

Ultrafast Molecular Frame Quantum Tomography

Luna Morigan¹, Simon P. Neville², Margaret Gregory^{1,*}, Andrey E. Boguslavskiy^{3,4}, Ruaridh Forbes^{3,5},
Iain Wilkinson^{2,6}, Rune Lausten², Albert Stolow^{2,3,4,7}, Michael S. Schuurman^{4,2},
Paul Hockett² and Varun Makhija^{1,†}

¹Department of Chemistry and Physics, University of Mary Washington, Fredericksburg, Virginia 22401, USA

²National Research Council Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada

³Department of Physics, University of Ottawa, 150 Louis Pasteur, Ottawa, Ontario K1N 6N5, Canada

⁴Department of Chemistry and Biomolecular Sciences, University of Ottawa, Ottawa, Ontario K1N 6N5, Canada

⁵Linac Coherent Light Source, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA

⁶Institute for Electronic Structure Dynamics, Helmholtz-Zentrum für Materialien und Energie,
Hahn-Meitner-Platz 1, 14109 Berlin, Germany

⁷NRC-uOttawa Joint Centre for Extreme and Quantum Photonics (JCEP), Ottawa, Ontario K1A 0R6, Canada

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We develop and experimentally demonstrate a methodology for a full molecular frame quantum tomography (MFQT) of dynamical polyatomic systems. We exemplify this approach through the complete characterization of an electronically nonadiabatic wave packet in ammonia (NH₃). The method exploits both energy and time-domain spectroscopic data, and yields the lab frame density matrix (LFDM) for the system, the elements of which are populations and coherences. The LFDM fully characterizes electronic and nuclear dynamics in the molecular frame, yielding the time- and orientation-angle dependent expectation values of any relevant operator. For example, the time-dependent molecular frame electronic probability density may be constructed, yielding information on electronic dynamics in the molecular frame. In NH₃, we observe that electronic coherences are induced by nuclear dynamics which nonadiabatically drive electronic motions (charge migration) in the molecular frame. Here, the nuclear dynamics are rotational and it is nonadiabatic Coriolis coupling which drives the coherences. Interestingly, the nuclear-driven electronic coherence is preserved over longer timescales. In general, MFQT can help quantify entanglement between electronic and nuclear degrees of freedom, and provide new routes to the study of ultrafast molecular dynamics, charge migration, quantum information processing, and optimal control schemes.

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Introduction.—Molecular quantum electronic dynamics [1–6] govern important natural processes, including photosynthesis [7], vision [8], photochemistry [9,10], and solar energy conversion [11]. Attosecond science probes population dynamics and coherences between electronic states [9,12–18]. The former often involves conical intersections generated by strong nonadiabatic coupling between the electrons and nuclei [1,6], the fundamental mechanism of energy transfer between them [10,19,20]. In general, the nuclear motions inducing such dynamics involve either rotation or vibration. Nuclear-driven electronic coherences generated at conical intersections are sensitive probes of their local topography [21–23]. Electronic coherences may play an important role in fundamental light-induced processes [7,12,24–27], thus measuring and controlling such coherences is of broad interest [18,28–38]. In general, measurement and control of electronic populations and coherences requires experimental determination of the time-dependent-electronic density matrix—a quantum tomography [39,40]. The latter underlies aspects of the

foundations of quantum mechanics [41–43] and molecular quantum information processing [44]. While probability distributions (static and dynamical) have been measured [45–50], quantum tomography was only demonstrated in restricted cases: a ground state rotational wave packet, a stationary vibrational state, and a dissociative vibrational state [51–55]. Recently, we proposed a systematic method for determination of the time-evolving electronic lab frame density matrix (LFDM) from experimental data [56]. We present here the first time-resolved molecular frame quantum tomography (MFQT).

Molecular frame quantum tomography in NH₃.—In this proof-of-concept demonstration, we resonantly excited a pair of electronic states in NH₃, nonadiabatically coupled by molecular frame (MF) rotation [57,58]. MFQT was achieved by combining data from ultrafast time-resolved measurements [58] with that of high-resolution spectroscopy [59]. The resulting density matrix yields the time-resolved electronic probability distribution in the MF, as a function of lab frame (LF) orientation angles. We show that

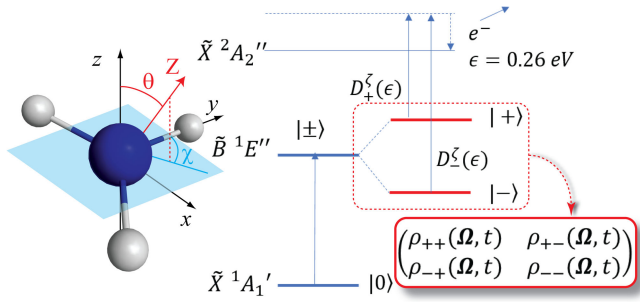


FIG. 1. Electronic coherences nonadiabatically driven by nuclear motion. NH_3 is excited to the \tilde{B}^1E'' state, a pair of near degenerate electronic states $|\pm\rangle$, the LF Z axis being the laser polarization direction. This excitation results in the time-dependent LF density matrix $\rho_{nn'}(\mathbf{\Omega}, t)$, with $n \rightarrow \pm$. The planar geometry of NH_3 is shown with MF symmetry axis z at angle θ with respect to the LF Z axis. In-plane rotation about z is given by the angle χ . Photoionization into the \tilde{X}^2A_2'' ionic state by the dipole operators $D_n^z(\epsilon)$ produces an electron with kinetic energy $\epsilon = 0.26$ eV. Both states ionize to overlapping continuum channels, ζ , permitting the detection of electronic coherences. As we show, rapid nuclear motion along the θ coordinate induces electronic coherences which drive charge migration in the MF. For details, see the text.

nuclear-driven charge distributions evolve differently at different MF orientations. Importantly, the observed aperiodic charge migration is direct evidence of an angle-dependent nonadiabatic coupling, the angular analog to vibrational-coordinate-dependent nonadiabatic coupling. In our example, the electronic coherence persists over the entire 5 ps window of the time-resolved experiment. Long-lived electronic coherences are rare [31,37], offering new opportunities for quantum control of molecular electronic dynamics [60], the study of electronic-nuclear entanglement [27,41,42], and the development of quantum information processing protocols in isolated molecules [44], for which quantum tomography is a necessary prerequisite.

In Fig. 1, we depict NH_3 resonantly excited from a thermal rotational state distribution in the ground \tilde{X}^1A_1' electronic state, $|0\rangle$, to its doubly degenerate \tilde{B}^1E'' state, $|\pm\rangle$, with three quanta in the umbrella vibrational mode. We determine the 2×2 orientation-dependent LFDM in the $|\pm\rangle$ basis, where $\Lambda_z|\pm\rangle = \pm|\pm\rangle$, and $L_z = \xi\Lambda_z$ [61]. L_z is the component of the electronic orbital angular momentum along the MF z axis, the threefold symmetry axis of NH_3 , and $\xi = \pm(\pm|L_z|\pm)$. In general, matrix elements of the LFDM can be written as [56]

$$\rho_{nn'}(\mathbf{\Omega}, t) = \sum_{KQS} A_{QS}^K(n, n'; t) D_{QS}^{K*}(\mathbf{\Omega}), \quad (1)$$

where $\mathbf{\Omega} = \{\phi, \theta, \chi\}$ are the MF Euler angles and $n, n' \rightarrow \pm$ indicate the coherently excited electronic states. Molecular angular distribution moments (MADMs)

$A_{QS}^K(n, n'; t)$ specify the evolving excited state molecular dynamics [56] and the $D_{QS}^K(\mathbf{\Omega})$ are Wigner D-Matrix elements [62]. MFQT is enabled by determination of all relevant MADMs from the experimental data. The MADMs are multipole moments of the LFDM which track the time varying anisotropy of each LF matrix element. Selection rules for linearly polarized light restrict us to MADMs with $K = 0, 2$, $Q = 0$ and $S = 0, \pm 2$, $|S| \leq K$ [56]. Furthermore, the symmetry of the \tilde{B}^1E'' state permits only three unique, nonzero MADMs: $A_{00}^0(+, +; t) = A_{00}^0(-, -; t)$, $A_{00}^2(+, +; t) = A_{00}^2(-, -; t)$ and $A_{02}^2(+, -; t) = A_{0-2}^2(-, +; t)$ [57,58,61]. The MADMs $A_{00}^0(\pm, \pm; t)$ track the total population in each excited state, while the $A_{00}^2(\pm, \pm; t)$ track the alignment of the z axis for the population in each state. The $A_{0\pm 2}^2(\pm, \mp; t)$ track the orientation of the electronic coherence in the lab frame. Their critical relationship to the electronic dynamics is detailed below.

Determining MADMs.—In both the time- and frequency-resolved experiments, the excited states were probed by single photon photoionization to the \tilde{X}^2A_2'' state of NH_3^+ [58,59,63,64]. The time-resolved data lead us to the time-dependent LFDM and the temporal evolution of the MF charge density. Here, NH_3 was excited by a 160.9 nm, 77 fs pump pulse, and ionized by a time delayed 400 nm, 40 fs probe pulse [58]. The photoelectron angular distribution and kinetic energy spectrum were measured as a function of time delay. Spherical harmonic expansion of the signal as a function of electron ejection angles θ_e and ϕ_e , $P(\theta_e, \phi_e, \epsilon, t) = \sum_{LM} \beta_{LM}(\epsilon, t) Y_{LM}(\theta_e, \phi_e)$, provides the time- and electron-kinetic-energy (ϵ)-dependent anisotropy parameters $\beta_{LM}(\epsilon, t)$. With linearly polarized pump and probe pulses, each a one-photon process, the three nonzero anisotropy parameters are β_{00} , β_{20} , and β_{40} [20,65,66]. These, in turn, are expressed in terms of the MADMs [20,67,68],

$$\beta_{LM}(\epsilon, t) = \sum_{KQS} \sum_{nn'} C_{KQS}^{LM}(n, n'; \epsilon) A_{QS}^K(n, n'; t). \quad (2)$$

Since the pump also generates three, unique nonzero MADMs then, with known coefficients $C_{KQS}^{LM}(n, n'; \epsilon)$, Equation (2) becomes a matrix equation with solution $\vec{A}(t) = \hat{C}^{-1} \vec{\beta}(t)$ at each time delay. NH_3 is well-studied spectroscopically [69–73]: the coefficients $C_{KQS}^{LM}(n, n'; \epsilon)$ comprising \hat{C} were previously determined by high-resolution resonant enhanced multiphoton ionization (REMPI) spectroscopy [59,63,64]. The coefficients can be written as $C_{KQS}^{LM}(n, n'; \epsilon) = \sum_{\zeta\zeta'} \Gamma_{KQS}^{\zeta\zeta'LM} d_{\zeta\zeta'}^{nn'}(\epsilon)$ with $d_{\zeta\zeta'}^{nn'}(\epsilon) = D_{\zeta}^n(\epsilon) D_{\zeta'}^{n'*}(\epsilon)$. The factors $\Gamma_{KQS}^{\zeta\zeta'LM}$ are analytical and their properties were previously discussed at length [74]. The $D_{\zeta}^n(\epsilon)$ are matrix elements of the dipole operator between the bound state labeled n and a continuum channel

ζ , specifying the final state of the ion plus free electron with kinetic energy ϵ . Some partial wave matrix elements were determined for several electron kinetic energies, constituting a “complete experiment” [67,68,75–79]. Here, we use the results for $\epsilon = 0.26$ eV, relevant to our time-resolved experiments. Equation (2) is valid for D_{3h} dipole matrix elements, symmetry adapted to the point group of NH_3 in its \tilde{B}^1E'' state. The $\vec{\beta}(t)$ from the time-resolved and \hat{C} from the frequency-resolved experiment (with associated experimental uncertainties) determine the MADMs [80]. The normalization $\sum_n A_{00}^0(n, n; t) = 1/8\pi^2$, equivalent to $\text{Tr}\{\rho(t)\} = 1$ [56], was applied at the initial time point and we rescaled the $K > 0$ MADMs such that the ratio A_{0S}^K/A_{00}^0 remains unchanged. The resulting MADMs track the time varying population and molecular orientation in each electronic state and, critically, the coherence between them. These construct the LFDM $\rho(\mathbf{\Omega}, t)$ in Eq. (1) for any MF orientation $\mathbf{\Omega}$.

Probing nonadiabatic dynamics.—We consider the electronic dynamics induced by nuclear motions. Selected elements of the extracted LFDM are plotted, at selected MF orientations, in Fig. 2. We note that the diagonal elements tracking populations, $\rho_{++}(\mathbf{\Omega}, t) = \rho_{--}(\mathbf{\Omega}, t)$, are the same for all three orientations (black crosses), increasing in the first 0.5 ps and then slowly decaying. This indicates a higher probability for MF orientations with $\theta = \pi/2$ after 0.5 ps independent of χ . The observed asymptotic behavior is expected for a perpendicular pump transition and a symmetric top geometry [57,69,86]. MF electronic dynamics at any orientation are dominated by the coherence $\rho_{+-}(\mathbf{\Omega}, t)$. The real part of $\rho_{+-}(\mathbf{\Omega}, t)$ is counterphased for molecules oriented with $\chi = 0$ (top) and $\chi = \pi/2$ (bottom), the imaginary part being zero. The electronic density at these orientations exhibit the complementary time evolution seen in the top and bottom rows. In contrast, at $\chi = \pi/4$ (middle) the real part of the coherence is zero, yielding entirely different electronic dynamics at this orientation.

From the orientation-dependent LFDM elements of Fig. 2, we construct the MF one-electron reduced density,

$$p(r_1, \mathbf{\Omega}, t) = \sum_{nn'} \rho_{nn'}(\mathbf{\Omega}, t) \int dr_2 \cdots dr_N \psi_n^*(\vec{r}) \psi_{n'}(\vec{r}), \quad (3)$$

where $\vec{r} = \{r_i | i = 1, 2, \dots, N\}$ is the set of position vectors of the electrons and $\psi_n(\vec{r})$ is the wave function for a basis state $|\pm\rangle$. This yields a one-electron attachment density, $p_A(\vec{r}_1, \mathbf{\Omega}, t)$, shown in Fig. 3, depicting the orientation- and time-dependent accumulation of MF electron density relative to the static reference ground electronic state (for details see Supplemental Material [80]). To be consistent with common usage, we will refer hereafter to the observed MF evolution of the attachment density as “charge migration” [18,31–36], but use this term to include both vectorial (directional) and tensorial (polarization)

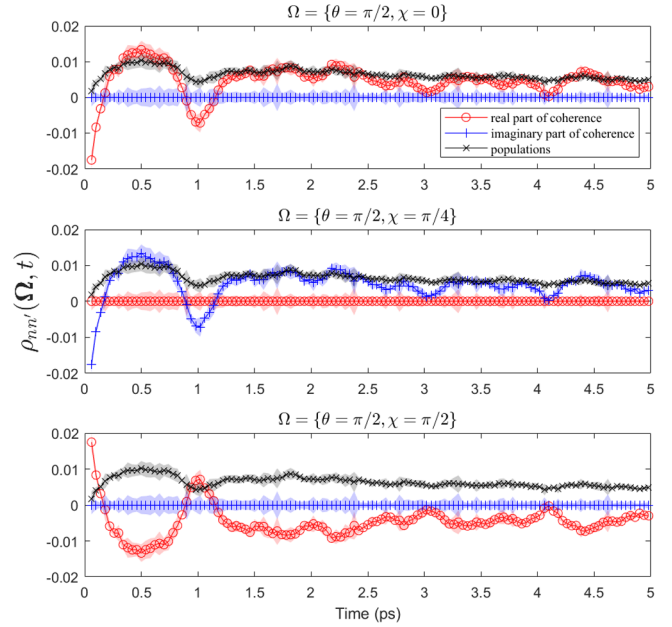


FIG. 2. Experimentally determined elements of the time-resolved LFDM, $\rho_{nn'}(\mathbf{\Omega}, t)$, for a molecule with z axis perpendicular to the laser polarization (i.e., $\theta = \pi/2$), for different in-plane rotation angles (see Fig. 1) $\chi = 0$ (top), $\pi/4$ (middle), and $\pi/2$ (bottom). The electronic populations $\rho_{\pm\pm}(\{\pi/2, \chi\}, t)$, black crosses, are independent of χ ; they initially increase then steadily decay, tracking the population of molecules oriented at $\theta = \pi/2$. In contrast, the electronic coherences $\rho_{+-}(\{\pi/2, \chi\}, t)$ vary with χ and are the dominant contribution to the charge migration dynamics. They are real but counterphased for $\chi = 0$ (top) and $\pi/2$ (bottom), indicating complementary electronic dynamics at these orientations. They are imaginary at $\chi = \pi/4$ (middle), revealing completely different electronic dynamics as a function of the nuclear coordinate χ .

moments of the electronic dynamics. The left and right columns show the attachment density migrating along the y axis for the orientations $\mathbf{\Omega} = \{\pi/2, 0\}$ and $\mathbf{\Omega} = \{\pi/2, \pi/2\}$, but in opposing directions. Comparing these with the coherences at $\mathbf{\Omega} = \{\pi/2, 0\}$ and $\mathbf{\Omega} = \{\pi/2, \pi/2\}$ of Fig. 2 reveals the correlation between the coherences and the MF electronic dynamics. At $\chi = 0$, as the coherence first increases (between 0 and 0.5 ps), the density migrates downward, reversing as the coherence subsequently decreases. Interestingly, at $\chi = \pi/4$, the one-electron attachment density migrates around the z axis. The radial extent of the plotted electronic density at all three orientations tracks the evolving population of perpendicularly oriented ($\theta = \pi/2$) molecules. A nuclear coordinate-dependent aperiodic migration of electronic density in the MF is direct evidence of non-adiabatic dynamics [15,85]. Since we excite a single vibrational state, the nuclear dynamics of relevance here are rotational. Using the LFDM, we construct the time-varying molecular axis distribution, $P(\theta, t) = \sum_n \rho_{nn}(\theta, t)$, plotted in Fig. 4(a), revealing the rotational dynamics, which are independent

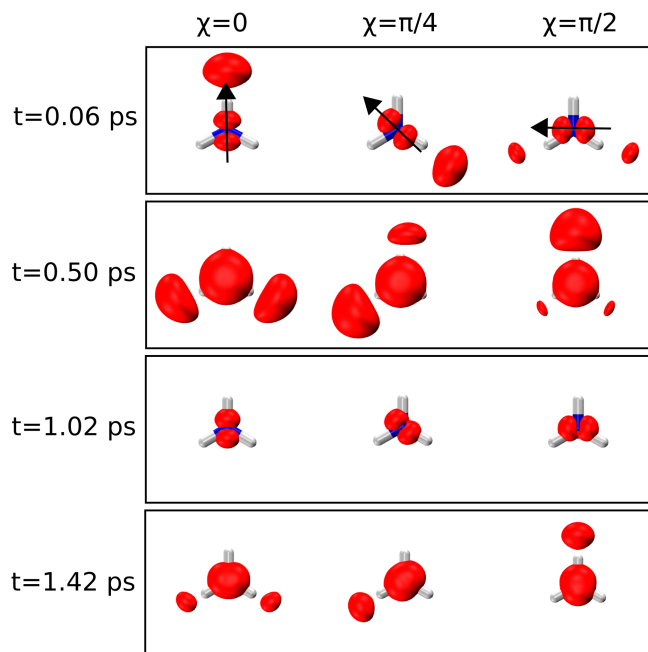


FIG. 3. Nuclear-driven MF charge migration, constructed from the experimentally determined $\rho(\mathbf{\Omega}, t)$. To illustrate, we show three columns depicting the time evolving attachment density $p_A(r_1, \mathbf{\Omega}, t)$ which tracks the variation of MF electron density, at three selected orientations; $\mathbf{\Omega} = \{\theta, \chi\} = \{\pi/2, 0\}$ (left), $\{\pi/2, \pi/4\}$ (middle), $\{\pi/2, \pi/2\}$ (right). The black arrow indicates the laser polarization direction Z (see Fig. 1). The electronic density evolves differently, and aperiodically, as a function of MF orientation, demonstrating nuclear coordinate-dependent nonadiabatic coupling between electronic states [15,85], the angular analog of the well-known vibrational-coordinate-dependent nonadiabatic coupling.

of χ as expected for a symmetric top [86]. In the first 1.5 ps, the most probable MF orientation oscillates rapidly between $\theta = 0$ and $\theta = \pi/2$. The electronic dynamics in Fig. 3 appear in this same time interval: the electronic coherence in Fig. 2 simultaneously exhibits large variations. Rapid nuclear motion, rotation of the MF z axis, at early times drives the electronic coherence and, therefore, the charge migration in the MF. The power spectrum of the electronic coherence is shown in Fig. 4(b), with peaks at $33.6 \pm 0.2 \text{ cm}^{-1}$ and its overtone, providing the timescale for MF charge migration: $0.99 \pm 0.3 \text{ ps}$. Later small fluctuations in $P(\theta, t)$ explain the persistent coherence. Beyond 1.5 ps, the most probable orientation remains relatively stable around $\theta = \pi/2$, with the time-averaged axis distribution peaking at $\theta = \pi/2$, as expected for a perpendicular transition [69,81,86]. Later frames of the MF electron density (see Supplemental Material [80]) show that the density remains localized, with only small fluctuations. Slower fluctuations of molecular alignment (slower MF θ rotation) at later times thus stabilizes the electronic coherence. Furthermore, due to Coriolis coupling, frequency components of the electronic coherence also appear in the

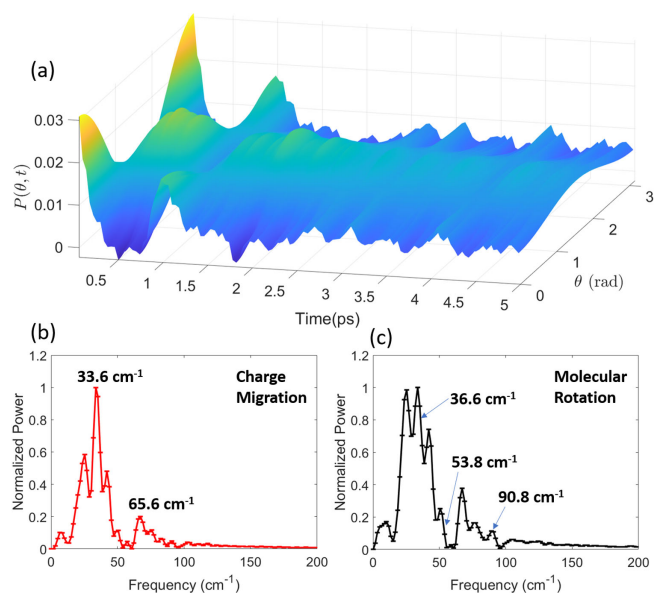


FIG. 4. Experimentally determined nuclear-driven electronic coherences in NH_3 . (a) The molecular Z axis distribution $P(\theta, t)$, determined from the experimental LFDM, characterizes the excited state rotational dynamics. It can be seen that the nuclear coordinate θ varies rapidly at first but slows down at later times; (b) Electronic coherences and charge migration. Power spectrum of the real part of the $\rho_{+-}(\{\pi/2, 0\}, t)$. The dominant frequency $33.6 \pm 0.2 \text{ cm}^{-1}$ (side bands at 25.2 ± 0.2 and $42.0 \pm 0.2 \text{ cm}^{-1}$). The overtone appears at $65.6 \pm 0.2 \text{ cm}^{-1}$. These determine the timescales of the nuclear-induced charge migration in the MF. (c) Rotational dynamics. Power spectrum of $P(0, t)$ with the locations of expected symmetric top rotational frequencies based on the deperturbed spectrum [81]. It can be seen that the quantum beats cannot be classified as either rotational or electronic, rendering the motions inseparable. See text for additional details.

power spectrum of $P(0, t)$, shown in Fig. 4(c). $P(0, t)$ also exhibits contributions, shown in Fig. 4(c), near expected locations of rotational quantum beats, determined assuming a symmetric top Hamiltonian [58,81]. Nevertheless, non-adiabatic coupling renders the electronic and rotational degrees of freedom non-separable. Therefore, all observed LF frequencies must be classified as quantum beats between ro-electronic molecular eigenstates. MFQT allows assignment of such beats by revealing which specific set of dynamical effects they contribute to in the LF and MF.

MFQT reveals the dynamics underlying nonadiabatic nuclear-driven electronic coherences. In this proof-of-concept example, the nuclear dynamics are rotational, with Coriolis coupling driving the nonadiabaticity [81]. Specifically, we note that (i) the rotational and electronic dynamics, separable in the cation-plus-free-electron final state [58], are nonseparable in the excited state; (ii) rapid rotation of the MF z axis (θ) at early times drives a dynamic MF charge migration with a $\sim 1 \text{ ps}$ period; (iii) subsequent small fluctuations of the MF z axis preserve the electronic coherence over a long time. We emphasize that all this

information is extracted from the experiment, without resorting to *ab initio* dynamical simulations [87,88].

Conclusions.—We conclude by considering limitations and future applications of MFQT to complex molecules, charge migration and quantum control, and foundational quantum mechanics in molecules. A clear limitation is that, in determining the LFDM, $\rho_{nn'}(\Omega, t) \equiv \langle \Omega n | \rho | n' \Omega \rangle$, we do not determine matrix elements of the density operator off-diagonal in the orientation angles, Ω . While this fully characterizes the electronic and vibrational dynamics in the MF, LF information is missing. We can construct the molecular axis distribution, but not observables sensitive to quantum coherences between different orientations, $\langle \Omega n | \rho | n' \Omega \rangle$, in the LF. Such observables are difficult to conceive since measurements relying on MF multipole interactions (like photoionization) are diagonal in $|\Omega\rangle$ by definition. The von Neumann entropy, $S = -\text{Tr}\{\rho \log \rho\}$, is one quantity containing these coherences and thus cannot be constructed here.

There remain important avenues of investigation. The entanglement entropy of the vibronic subsystem, $S_{vib}(t) = -\text{Tr}\{\tilde{\rho}(t) \log \tilde{\rho}(t)\}$, where $\tilde{\rho}_{nn'}(t) = 8\pi^2 A_{00}^0(n, n'; t)$ is the reduced vibronic density matrix, can be constructed. The time-varying electron entropy, $S_{el}(t)$ in the $\text{NH}_3 \tilde{B}^1 E''$ state may provide a quantitative measure of the electronic-rotational entanglement [89–92]: its time dependence may illuminate the role of entanglement in molecular electronic dynamics [41–43,93]. Investigating entanglement with an initially thermalized subsystem, as in this example, is an interesting prospect from the perspective of quantum thermodynamics [89,92,94]. Opportunities for optimal quantum control of MF dynamics via $\rho(\Omega, t)$ also emerge [60]. For instance, in NH_3 , the $|\pm\rangle$ states may be controlled by the non-resonant dynamic Stark effect [95–97]. Manipulating the LFDM in such a manner would control the time dependence of the electron density, a feature directly relevant to the burgeoning field of ultrafast molecular chirality [98,99]. MFQT would allow similar experimental manipulation of charge migration in molecules, since the MF charge dynamics are directly accessible experimentally.

Photoionization-based MFQT requires as input complete REMPI experiments achieved only for a handful of molecules [59,63,64,75]. Emerging attosecond techniques may be applicable: rotational wave packet studies [100] or angle-resolved RABBIT [101,102] may provide sufficient information for *in situ* complete photoionization experiments from an electronic molecular wave packet. In general, when many electronic and/or vibrational states are excited, the matrix inversion problem in Eq. (2) becomes ill posed. Sophisticated mathematical methods were developed to deal with such situations, if physical constraints can be provided [82,103]. Although the complete photoionization experiment problem itself can be similarly ill posed, only products of the dipole matrix

elements are needed to determine \hat{C} , circumventing the more complex problem of determining individual dipole matrix elements [74]. High quality *ab initio* dipole matrix elements [104–107] may provide another suitable methodology. Finally, other angle-resolved scattering probes sensitive to the MADMs also apply [108–110], provided the link between experiment and the MADMs is rigorously determined. We anticipate that this work will inspire a number of interesting directions in the study of quantum dynamics, charge migration, coherences, and entanglement in isolated molecules.

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*Current address: Department of Earth, Atmospheric, and Planetary Sciences, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA.

†vmakhija@umw.edu

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