



The Ultrafast Structural Dynamics of Ammonia Cation Following Strong-Field Ionization

Beer Bahadur Singh^{1*}, Dr. K K Singh²

1. Research Scholar, F S University, Shikohabad, Uttar Pradesh, India
beeruchaudhary@gmail.com ,

2. Assistant Professor, Department of Physics, F S University, Shikohabad, Uttar Pradesh, India

Abstract: This work examines rapidly changing structural dynamics of ammonia cation (Nha^+) after strong field ionization by use of Fourier transform laser-induced electron diffraction (FT-LIED). Through the analysis of high-energy electrons backscatter in the rescattering regime, molecular interference signals were obtained and converted to extract real-space structural information without using theoretical fitting models. The outcome of the results shows that the internuclear distances are $.31 + 0.03$ Å (N–H) and $2.24 + 0.03$ Å (H–H), which result in H - N - H bond angle of $117 + 5$ degree. Complementary quantum chemistry simulations show the strong laser field has a significant effect on the potential energy surface due to which is a field-dressed structure that is consistent with experimental measurements. These results demonstrate that FT-LIED is capable of probing the geometry of molecules directly in the femtosecond time range and can give information regarding the contribution of strong-field effects to the mechanisms of structural changes in polyatomic molecules.

Keywords: Ammonia Cation, Strong-Field Ionization, Ultrafast Structural Dynamics, Fourier Transform, Polyatomic Molecules

----- X -----

INTRODUCTION

The study of ultrafast molecular dynamics has emerged as one of the key goals in the current chemical physics, especially in strong-field laser-matter interactions. When the molecules are subjected to high femtosecond laser pulses, they are quickly ionized and the electron-nuclear dynamics develop quickly on the sub-femtosecond to femtosecond scales. These mechanisms are the ones that determine the basic phenomena like breaking of bonds, redistribution of charges and even structural rearrangement. The recently developed methods of ultrafast spectroscopy and imaging have facilitated the observation of these short-lived states directly, and this has provided new insights into the behavior of the molecules well out of equilibrium state as has never been known before. [1][2]

In comparison to other molecular systems, ammonia (NH_3) is considered to be an archetypal polyatomic molecule due to the fact that it has the well-known electronic structure and a characteristic inversion motion, also known as an umbrella motion. The cationic structure may undergo a potential energy surface modification, which can result in a quick transformation of the neutral pyramidal structure of ammonia into a planar structure. This transformation can take place following ionization. It has been established both experimentally and theoretically that such a transition from pyramidal to planar is achieved on a very short timeframe of a few femtoseconds. As a result, ammonia is an excellent system for the study of real-time structural dynamics. It is possible to employ these high-rate geometric changes as a test bed for higher order time-resolved experimental approaches.

When it comes to the beginning of these explosive events, active ionization in the strong field is absolutely necessary. It is possible for electrons to tunnel ionize when the laser intensity is strong. It is then possible for the electrons to be repulsed by the vibrating electric field to the parent ion, which results in rescattering. When the recollision takes place, the information that is contained inside these dispersed electrons is the structural information of the parent molecule. Laser-induced electron diffraction (LIED), which has been developed into a powerful tool for visualizing molecular structures on an Angstrom spatial and femtosecond temporal scale, is based on this phenomenon, which serves as its basis. It is [3]. In addition, the fact that LIED is capable of directly investigating molecules that include hydrogen contributes to the significance of this technique in the field of light-atom dynamics research.

The techniques of LIED have, up until relatively recently, been subjected to the use of Fourier transform LIED (FT-LIED), which has given rise to the possibility of retrieving the structure of molecules in experimental data via the utilization of model-independent approaches. As contrast to traditional approaches that are dependent on a theoretical fit, FT-LIED is used to directly identify structural information by making use of observed electron momentum distributions. A far more exact and trustworthy structural determination is now possible as a result of this innovation, especially with regard to the formation of transient strong-field molecular states. [4] As a consequence of this, FT-LIED has become an essential tool for the investigation of the fast structural change of molecules while they are in the gas phase.

In addition to the development of experiments, theoretical research have placed an emphasis on the function that surfaces of field-dressed potential energy play in the study of molecular dynamics in powerful laser fields. It is possible for strong electric fields to have a significant influence on the potentials of molecules, which may result in equilibrium geometries and reaction pathways being disrupted. These kinds of field effects are particularly important in systems like ammonia, where the structure of the movements that are seen is determined by the equilibrium between the nuclear motion and the electronic motion. The most current computational study highlights the fact that these field effects should not be ignored since they often lead to interpretations of experimental results that are either inadequate or incorrect. [5][6]

Moreover, the study of the dynamics of ionization in ammonia and related systems has shown a complicated mechanism which includes the mobility of charges, the transfer of protons, and the nonadiabatic interactions. They are very fast, taking place on a timescale of orders of magnitude, and very sensitive to electronic excitation and molecular environment. The research on ammonia clusters and dimers has shown that ionization can cause a rapid redistribution of energy, as well as structural rearrangement, indicating the complex interplay between the dynamics of electrons and the dynamics of nucleus. [7] Experimental techniques with the ability to capture these coupled dynamics in real time are important because of such findings.

Here, the current research is aimed at the ultrafast structural evolution of the ammonia cation (NH_3^+) after ionizing it in strong-field under FT-LIED. Through the analysis of backscattered high-energy electrons, we obtain the transient molecular geometry in a direct manner, not using theoretical modeling. The findings are a clear indication of the presence of a near-planar field-dressed structure and give novel insights into the importance of strong laser fields in evoking structural changes. The article is part of the general knowledge about ultrafast molecular imaging and defines FT-LIED as an effective method of study of real-time

dynamics in polyatomic molecules. [8]

OBJECTIVES

- To investigate the ultrafast structural dynamics of NH_3^+ following strong-field ionization.
- To retrieve molecular geometry using the FT-LIED technique without theoretical modeling.
- To analyze the influence of strong laser fields on molecular structure using quantum chemical calculations.

METHODOLOGY

Experimental Configuration

A home-built mid-infrared optical parametric chirped-pulse amplifier (OPCPA) device that could generate ultrashort laser pulses suitable for strong-field ionization study was employed for the experimental investigations. With a central wavelength of 3.2 μm , a pulse duration of 100 fs (full-width at half maximum), and a repetition rate of 160 kHz, this system produced laser pulses that essentially offset the decreased rescattering cross-section of long wavelength regimes at a scaling factor of 3.2⁴. An on-axis paraboloid mirror installed in the reaction microscope chamber was utilized to concentrate the laser beam and turn it into a supersonic molecular beam. A maximum laser intensity (I_0) of 1.3×10^{11} W/cm² was reached with a focused spot size of about 67.8 μm . With a maximum classical return energy of around 380 eV ($3.17U_p$) and a maximum scattered electron energy of roughly 1200 eV ($10U_p$), this high intensity results in a ponderomotive energy (U_p) of 120 eV, which characterizes the dynamics in the electron population in the laser field. Furthermore, the predicted Keldysh value ($\gamma = 0.2$) indicates that the ionization process occurs mostly in the tunneling domain, demonstrating the applicability of strong-field theoretical models for further research.

Detection and Reaction Microscope

A reaction microscope (ReMi) was used to detect charged particles created during the contact, which made it possible to analyze coincidences with high precision as long as the experiment was carried out in ultrahigh vacuum. A cold molecular beam of ammonia 5% NH_3 in 95% helium was introduced into the contact area to achieve supersonic expansion, resulting in a high target density and little thermal broadening. Ionization events and fragmentation occurred when they were allowed to mix with the centered laser pulses. The resulting ions and electrons were then directed toward two opposing MCP detectors with position-sensitive delay-line anodes by homogeneous electric and magnetic fields of 34 V/cm and 13 G, respectively. Using time-of-flight (ToF) measurements along the spectrometer axis and spatial impact locations on the detector plane, the system made it possible to estimate the three-dimensional momentum vectors (p_x , p_y , and p_z) of the particles identified with high precision. This was made feasible by the complete electron-ion coincidence detection capacity, which made it possible to identify specific reaction channels and accurately correlate ionization dynamics with the ensuing molecular fragmentation paths.

LIED-Based Structural Retrieval

The structural dynamics of the ammonia cation were observed using LIED, a strong-field technique based

on the rescattered electrons as molecular structure probes on the femtosecond scale. Elastic scattering of the parent ion and momentum changes caused by the vector potential of the laser field just before to rescattering make up the observed distributions of electron momentum using this approach. The classical recollision model provides a reliable framework for tracking the electron tracks in quasi-static tunneling and accurately depicts the dynamical evolution of the vector potential. In this study, the vector potential was computed using distinctive aspects of the acquired electron spectra, which included the high-energy cutoff at $10U_p$ and the transition area between direct and rescattered electrons at $2U_p$. Using the formula $U_p = E_0^2 / 4\omega^2$, the ponderomotive energy was proportional to the electric field's amplitude. Additionally, the ADK ionization theory, which was used to match the longitudinal momentum distribution of the Ar⁺ ions, produced laser peak intensity measurements that were equal to the experimentally reported value of 1.3×10^{11} W/cm². The rescattering process was recreated by tracing the final measured electron momentum parallel and perpendicular to the laser polarization axis in relation to the return momentum and scattering angle. Fourier transform LIED enabled the direct extraction of structural information in the case of backscattered electrons ($\theta_r = 180^\circ$) after the correction of the laser field's influence. Long electron trajectories predominate due to their higher ionization potential, as demonstrated by classical trajectory analysis. As a result, they were primarily considered during the reconstruction process, which occasionally allowed the measurement of momentum distributions to provide a direct correlation with the time-dependent change in the molecular structure.

Computational Methodology

Quantum chemical calculations were performed to determine adiabatic ground-state PES along the inversion coordinate of both neutral NH₃ ($X_1^1A_1$) and its cationic analog NH₃⁺ (\bar{X}^2A_2) in order to further complement the experimental results and provide more detailed information on the structural development of the ammonia cation. To ensure an accurate representation of electron correlation, these calculations were performed using the CCSD technique with the aid of the Q-Chem 5.1 computer program and the augmented correlation-consistent polarized valence double-zeta (aug-cc-pVDZ) as a basis set. At each point of the PES, the permanent dipole moments and dipole polarizabilities in the presence of a static electric field were estimated. The molecular system's interaction with an external electric field with a strength of 0.06 atomic units the conditions in the experiment under the influence of the laser was then incorporated to obtain the field-dressed energies. Using the ANO-RCC-PVDZ basis set in the openMolcas 8.0 computational tool, second-order Moller-Plesset (MP2) theory was used to improve the initial molecular geometries. A dummy atom was placed along the molecule's primary axis of symmetry in order to determine the inversion coordinate. The bond angles between the H and N as well as X were purposefully altered between 130° and 90° in increments of 10° in all 41 feasible geometrical configurations. The planar geometry ($Q = 0^\circ$), which is a bond angle of 120°, was used to create the inversion coordinate (Q). The value of (Q) is either positive or negative depending on how far the nitrogen atom is from the hydrogen atom plane. The entire description of the inversion dynamics and the crucial support for the interpretation of the experimentally observed ultrafast dynamics of the structural change in the ammonia cation were made possible by this computational model.

RESULTS

FT-LIED Analysis

The FT variant of LIED, sometimes referred to as FT-LIED and fixed-angle broadband laser-driven electron scattering (FABLES), is used in this work to retrieve molecule structural information. Here, a Fourier treatment eliminates a coherent molecular interference signal (q_m) within the momentum distribution of extremely high-energy backscattered electrons ($\theta_r = 180^\circ$) to provide a real-space representation of the molecular structure in the far-field. The primary advantage of the FT-LIED technique is that the total signal of detected interference (q_e) includes the experimentally determined incoherent background contribution due to atomic scattering (q_a). The molecular interference term (q_m) may be derived immediately by extracting this experimentally observed background and subtracting it from the overall signal. This allows for structure recovery without the need for complex modeling techniques, retrieval algorithms, or fitting theories.

The electrons' coincident momentum distribution is a logarithmically scaled distribution of electrons found in relation to the transverse momentum (P_Ψ), which is perpendicular to the laser polarization, and the longitudinal momentum (P_\parallel), which is parallel to it. This pattern clearly distinguishes between the direct and rescattered electron contributions. Direct electrons are pushed to gather velocity primarily by the vector potential of the laser field at the ionization site and are lost from the parent ion without rescattering. As a result, their longitudinal momentum is about $P_\parallel \rightarrow \leq 4.2$ a.u., and their maximal kinetic energy is $2U_p$. You. The pulsating laser field pushes the rescattered electrons back toward the parent ion, where they undergo elastic scattering and acquire much more kinetic energy. At the instant of rescattering, these electrons have a return momentum (k_r) and are also kicked by the laser field's vector potential $A(t_r)$. Accordingly, $k_{\text{resc}} = k_r + A(t_r)$ yields the observed final momentum (k_{resc}). Since the rescattered electrons' peak accelerating kinetic energy may reach $10U_p$, their longitudinal momentum is about $P_\parallel = 9.4$ a.u. In the momentum window, $2U_p \leq E_{\text{resc}} \leq 10U_p$ (i.e., $4.2 \leq P_\parallel^2 \leq 9.4$ a.u.), this strong energy differential allows the elastic rescattering electrons (i.e., the structural information) to be clearly distinguished from the direct electrons.

Only electrons that meet certain selection criteria may be tallied in the FT-LIED analysis in order to obtain the proper structural retrieval. More precisely, low-energy direct electrons that offer no structural information are essentially eliminated by selecting coincidence electrons with a return momentum $k_r > 2.1$ a.u. (equivalent to $P_\parallel^2 > 4.2$ a.u.). Additionally, it is restricted to electrons dispersed across a small angular range around the backscattering condition ($\theta_r = 180^\circ$), with angular errors ($\Delta\theta_r$) ranging from 2 to 10 o. Smaller angle windows may be employed at higher momenta since direct electrons are negligible due to their lower energies, while greater angular ranges are allowed for lower return momenta to lessen the contribution of direct electrons. By integrating throughout a designated region in momentum space, often represented as a block arc, at different values of the vector potential shift (A_r), the signal resulting from the molecular interference is collected. This careful selection and integration technique can provide the high-quality interference signals required for the Fourier transform analysis and enable a precise reconstruction of the ammonia cation's molecular structure.

B. Electron-Ion 3D Coincidence Identification

It is possible that investigations using strong-field laser-induced electron diffraction (LIED) might entail additional phenomena in addition to the elastic scattering of tunnel-ionized electrons. As an example, it is possible for several ionizations to take place, which would result in the removal of more than one electron from the molecule. This results in the Coulomb explosion of multiply charged NH_3^{n+} , which in turn leads to the emergence of fragment ions such as NH_2^+ and H^+ , along with the electrons that correspond to them. There is also the possibility that background molecules in the main chamber might be a contributor. These molecules can generate species of ions that are not of relevance to this research, such as H_2O^+ , N_2O^+ or O_2 .

The whole collection of these background ions, together with the electrons that are associated with them, are viewed in the spectrometer alongside the molecular ion of interest, which is NH_2^+ . The ion that is found to be the most prominent in the ion time-of-flight (ToF) spectrum (Figure 1(a)) is the one that is observed to be at around $4.1 \mu\text{s}$. In the FT-LIED analysis, background ion electrons produce noise that is not acceptable, and as a result, they inhibit the accurate recovery of structures when the signals are averaged across all of the ionization channels.

The electron-ion coincidence detection technique is applied in order to ensure that the LIED interference signal is generated only by the ion of interest (NH_3^+). This technique is employed in order to address the difficulty. It is possible to understand the significance of this approach by referring to Figure 3(b), which shows the overall signal of electrons on all ions (in the color petrol blue) in comparison to electrons recorded in coincidence with NH_2 (in the color orange). The traditional cutoffs at $2U_p$ and $10U_p$ are visible in both distributions, and they are represented by dashed lines that run vertically across the graph.

Significant (an order of magnitude) decrease in the number of electrons in the rescattering region is seen in case of coincidence detection. Moreover, inset in Figure 1(b) indicates stronger oscillations in the coincidence-selected distribution (orange) which are caused by the molecular interference signal. These oscillations are very clearer than the total electron distribution oscillations (petrol blue), which supports the fact that coincidence detection is effective in improving the quality of structural signals.

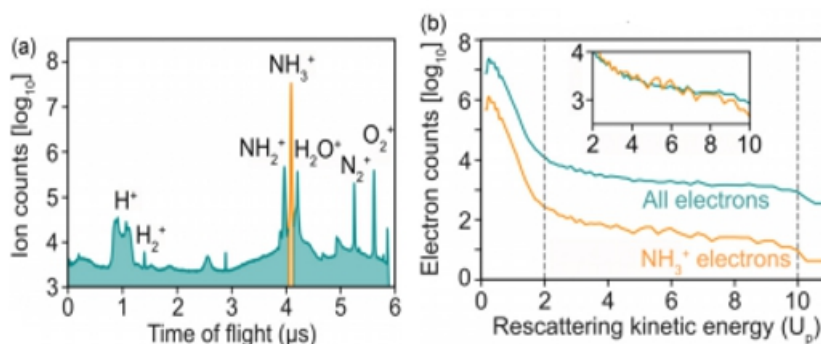


Figure 1: Electron-ion coincidence detection.

1(a) Ion ToF spectrum peaks at 4.1 μs , indicating NH_3^+ .

1(b) Electron signal vs. rescattered kinetic energy (U_p) for all electrons and those coincident with NH_3^+ . The 2-10 U_p area is shown in the inset, with the NH_3^+ distribution scaled by 50.

C. Retrieval of Molecular Structure

The rescattering plateau ($2U_p$ – $10U_p$) is defined as the distribution of return kinetic energy 40–350 eV, and it is used to quantify coincidence electrons with NH_3^+ ions. A background signal (q_β) that is unrelated to molecule structure and consists of the incoherent sum of atomic scatterings, and a coherent signal (q_m) that is a result of molecular interference make up the observed backscattered electron distribution (q_e). In order to get the structural details, we need to get the interference signal, which is derived by subtracting a known background using a third-order polynomial fit of the logarithmic electron distribution, expressed as $q_m = \log_{10}(q_e/q_\beta)$. In the back-rescattered frame, the observed oscillatory behavior provides a sensitive and unique indication of the underlying molecular structure, which is subsequently described as the interference signal as a function of the momentum transfer ($q = 2k_r$).

Additionally, real-space structural characteristics are obtained from analyses of the molecular interference pattern by means of a fast fourier transform (FFT). The use of zero padding and a Kaiser window ($\beta = 0$) prior to transformation improves spectral resolution and reduces the impact of edge effects. There are two distinct peaks in the resulting FFT spectrum, individual Gaussian fits, and their combined profile, and they are all attributed to distinct internuclear separations. Two peaks can be seen, one at $1.31 \pm 0.03 \text{ \AA}$ and the other at $2.24 \pm 0.03 \text{ \AA}$. The reported structural characteristics, including the former NH internuclear distance (R_{NH}) and the later H-H internuclear distance (R_{HH}), are consistent with these values. The presence and positioning of these peaks confirm the reliability of the FT-LIED method in instantly establishing the molecular geometry via experimentation.

The H-N-H bond angle, which is $117 \pm 5^\circ$ in accordance with the measured internuclear distances, is a geometrical reflection of the ammonia cation. When these results are compared to the previously published values of neutral NH_3 and the cationic forms of this acid, it provides more evidence that these findings are legitimate. Properties beyond 3 \AA are anticipated to be caused by the presence of ammonia clusters, which are recognized to have center-of-mass (N-N) distances ranging from 3.2 to 5.2 \AA and lengthy H-H spacing lengths of 8 \AA . These results show that the method can find the internal structure of molecules and also imply that it can find any intermolecular interaction at room temperature.

D. Calculations in quantum chemistry.

Quantal chemical ab initio calculations have been performed to study the pyramidal-planar geometrical transformation of ammonia up to the point of strong-field ionization in order to help understand the NH_3^+ structure that was observed by FT-LIED. The neutral NH_3 and NH_3^+ cations' ground-state potential energy curves (PECs) are shown in Figure 5, with the former shown as black solid curves and the latter as colored dashed curves, indicating field dressing. Figure 5 shows that the inversion coordinate (Q) is zero, which indicates the planar geometry; negative and positive values, on the other hand, show that the nitrogen atom is above and below the plane of the three hydrogen atoms, respectively, and hence determine the molecular geometry. Change in structure at the instant of ionization ($t = 0 \text{ fs}$), the structural transition is initiated when

a neutral ammonia nuclear wave packet (NWP) is projected onto the potential energy surface of the NH_3^+ cation. When the NWP is operated under field-free circumstances, the planar equilibrium geometry ($Q = 0$; $\angle\text{H-N-H} = 120^\circ$) is achieved in about 7.9 fs. Experimentally, near-planar geometries may be probed with MIR-LIED since this period is comparable to the 7.8 – 9.8 fs required to return the LIED electron to the parent ion.

In order to overcome these shortcomings, field-dressed Born-Oppenheimer potential energy curves were computed at a field strength at which the experiment achieved peak intensity ($1.3 \times 10^{14} \text{ W/cm}^2$, which is also 3.1 V/Å) as demonstrated in Figure 5. The outcomes show that the presence of the powerful laser field alters the geometry of equilibrium of the cation by pushing the geometry away of the planar structure to the bent one ($Q \approx 14$; $\angle\text{H-N-H} \approx 114^\circ$). The field-dressed curves also show that the response of the molecules is dependent on the orientation of the laser polarization with the dipole moment as indicated by the orange and blue dashed curves depicting parallel and antiparallel arrangement of the polarization and the dipole moment respectively. It is interesting to note that the experimentally derived FT-LIED bond angle ($117 \pm 5^\circ$) and the theoretical equilibrium geometry of the field-dressed ground cationic state ($\approx 114^\circ$), are in strong agreement ($117 \pm 5^\circ$). Though the current calculation gives useful static analysis, the ultimate comprehension of the ultrafast evolution of structure will be time dependent using quantum dynamical calculations that are suggested to be employed in the future research.

Figure 5 shows the field-dressed Born-Oppenheimer potential energy curves that were estimated to overcome these constraints. The field strength used was $1.3 \times 10^{14} \text{ W/cm}^2$, which is the same as 3.1 V/Å. The findings show that the cation's equilibrium geometry changes due to the intense laser field, moving it from a flat structure to a bent one ($Q \approx 14$; $\angle\text{H-N-H} = 114^\circ$). The molecular response is also influenced by the alignment of the laser polarization with regard to the dipole moment, as seen by the field-dressed curves for parallel and antiparallel orientations, respectively, indicated by orange and blue dashed curves. Significantly, the computed equilibrium geometry of the field-dressed ground cationic state ($\approx 114^\circ$) and the experimentally determined FT-LIED bond angle ($117 \pm 5^\circ$) well coincide. Despite the useful static insights provided by the current computations, future research is encouraged to use time-dependent quantum dynamical simulations in order to fully comprehend the ultrafast structure development.

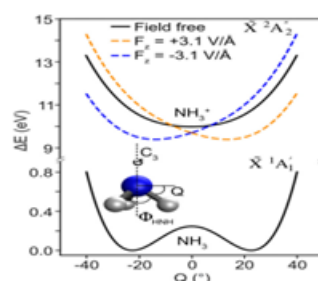


Figure 2: Quantum chemistry calculations, Field-free and field-dressed potential energy curves of NH_3 and NH_3^+ along the inversion coordinate, showing dipole-aligned field effects and molecular geometry.

It has been shown in the current research that with Fourier transform laser induced electron diffraction (FT-LIED), the molecular structure of the ammonia cation can be accurately and directly retrieved after strong-

field ionization. Backscattered electron momentum distributions were analyzed in the rescattering regime ($2U_p-10U_p$) to demonstrate clear patterns of interference, and internuclear distances of $1.31 \pm 0.03 \text{ \AA}$ (N-H) and $2.24 \pm 0.03 \text{ \AA}$ (H-H) were obtained, which would yield an H-N-H bond angle of $117 \pm 5^\circ$. These observations suggest that the NH_3^+ molecule assumes an almost planar geometry at the ultrafast timescale, measured by the returning electron wave packet. Further Complementary quantum chemistry calculations also verified that the strong laser field alters the potential energy surface and the equilibrium structure is no longer a planar structure but a slightly bent structure ($\sim 114^\circ$) which is very consistent with the experimental results. In general, the findings demonstrate the potential of FT-LIED to resolve the ultrafast structural dynamics of high spatial and temporal sensitivity, as well as the high importance of field-induced effects in governing molecular geometry during strong-field ionization processes.

DISCUSSION

The current results emphasize the usefulness of Fourier transform laser-induced electron diffraction (FT-LIED) to study the dynamics of the structure of the ammonia cation at high temporal and spatial resolution. The experimentally measured near-planar geometry (H-N-H $\approx 117 \pm 5^\circ$) suggests structural reorganization to be taking place on the timescale of a few femtoseconds after strong-field ionization as expected of electron rescattering. The fact that it is not entirely planar indicates the strong laser field has a significant effect on the potential energy surface, and causes field-dressed molecular structures. This finding is in line with the current research which focuses on the importance of laser-matter interaction in modifying the structure of the molecules during ionization and rescattering process. [9] [10] Moreover, experimental results of bond parameters and field-dressed quantum chemical calculations of the measurements of the same bonds confirm that FT-LIED is a reliable structural probe that is independent of the model. [11] The fact that high-energy rescattered electrons, in the regime of $2U_p-10U_p$, have a contribution to the sensitivity to internuclear distances supports previous reports of the capability of LIED methods to resolve at the level of Angstrom. [12] Moreover, the existence of the weak signals in the larger internuclear distances indicates the likelihood of the existence of clustering effects that have been reported under similar experimental conditions and could have played some roles in the observed diffraction patterns. [13] In general, the research highlights the significance of integrating field effects and ultrafast electron dynamics in order to understand molecular structure correctly in strong-field regimes.

CONCLUSION

Finally, the current study makes FT-LIED an effective and model-free method of imaging high spatial and time resolution ultrafast molecular structural dynamics. The experimental findings prove the idea that ammonia experiences a rapid pyramidal-to-near-planar transition in the post-ionization process, and the experimentally measured structural parameters are in close agreement with field-dressed quantum chemical calculations. The paper also highlights the powerful effect that the great laser field has in molecular geometry that should be taken into account in the proper interpretation of the structure. Also, capacity to separate coherent molecular interference with background scattering improves reliability of the structural retrieval. Altogether, this article improves the ongoing body of ultrafast molecular imaging and paves the way to the investigation of real-time alterations in structural properties of complex molecular systems under the strong-field conditions.

References

1. Belsa, B., Amini, K., Liu, X., Sanchez, A., Steinle, T., Steinmetzer, J., ... & Biegert, J. (2021). Laser-induced electron diffraction of the ultrafast umbrella motion in ammonia. *Structural Dynamics*, 8(1).
2. Kraus, P. M., & Wörner, H. J. (2018). Perspectives of attosecond spectroscopy for the understanding of fundamental electron correlations. *Angewandte Chemie International Edition*, 57(19), 5228-5247.
3. Zhou, J., Wu, L., Belina, M., Skitnevskaya, A. D., Jia, S., Xue, X., ... & Ren, X. (2025). State-and time-resolved observation of ultrafast intermolecular proton transfer in hydrated biomolecules. *Nature Communications*, 16(1), 5838.
4. Huang, X., Lei, K., Mi, Y., Fang, W., & Li, X. (2023). Recent progress on hydrogen production from ammonia decomposition: technical roadmap and catalytic mechanism. *Molecules*, 28(13), 5245.
5. Humphreys, J., & Tao, S. (2024). Advancements in Green Ammonia Production and Utilisation Technologies. *Johnson Matthey Technology Review*, 68(2).
6. Li, S., Zhou, Y., Fu, X., Pedersen, J. B., Saccoccio, M., Andersen, S. Z., ... & Chorkendorff, I. (2024). Long-term continuous ammonia electrosynthesis. *Nature*, 629(8010), 92-97.
7. Wang, X., Tsimpidi, A. P., Luo, Z., Steil, B., Pozzer, A., Lelieveld, J., & Karydis, V. A. (2025). The influence of ammonia emissions on the size-resolved global atmospheric aerosol composition and acidity. *EGU sphere*, 2025, 1-35.
8. Tao, Y., Lei, L., Liu, J., & Wu, B. (2025). Local Proton Disorder Induced Intermolecular HH Coupling in Ionization of Dense Ammonia. *arXiv preprint arXiv:2510.17706*.
9. Pullen, M. G., Wolter, B., Le, A. T., Baudisch, M., Hemmer, M., Senftleben, A., ... & Biegert, J. (2015). Imaging an aligned polyatomic molecule with laser-induced electron diffraction. *Nature Communications*, 6(1), 7262.
10. Chirvi, K., & Biegert, J. (2024). Laser-induced electron diffraction: Imaging of a single gas-phase molecular structure with one of its own electrons. *Structural Dynamics*, 11(4).
11. Zhang, M., Guo, Z., Mi, X., Li, Z., & Liu, Y. (2022). Ultrafast imaging of molecular dynamics using ultrafast low-frequency lasers, X-ray free electron lasers, and electron pulses. *The Journal of Physical Chemistry Letters*, 13(7), 1668-1680.
12. He, L., Zhu, X., Cao, W., Lan, P., & Lu, P. (2022). Attosecond spectroscopy for filming the ultrafast movies of atoms, molecules and solids. *Chinese Physics B*, 31(12), 123301.
13. Tuthill, D. R. (2023). *Attosecond Probes of Molecular Electron Dynamics*. The Ohio State University.