

High-Resolution Spectroscopic Characterization of the ν_2 saQ(6, 3) Rovibrational Transition of Ammonia, an Important Air-Quality Pollutant, Using an Ultra-Stable Laser Spectrometer

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Abstract

This paper reports a high-resolution spectroscopic study of the ν_2 saQ(6, 3) rovibrational transition of ammonia (NH_3) near 10 μm , performed with an ultra-stable laser. Forty-six absorption spectra were recorded at 300 K and 10 Pa and processed with an automated non-linear fitting procedure implemented in Python. Two line-shape models were compared: a Gaussian profile, accounting for Doppler broadening only, and a Voigt profile, obtained from the convolution of the Doppler and collisional (Lorentzian) contributions. The Gaussian model converged for all 46 spectra and provided a numerically robust estimate of the transition frequency, whereas the Voigt model, although physically more complete, converged for only 22 of the 46 spectra, reflecting its greater sensitivity to the signal-to-noise ratio and to baseline quality in the wings of the line. Residual analysis shows that a purely Doppler profile does not fully reproduce the observed line shape, while Voigt residuals remain structured for several spectra with lower spectral quality. The central frequency obtained in this work, 28,953,694.0613 MHz, is consistent with the range of previous experimental determinations of this transition and falls within about 1 MHz of the ab initio prediction, with the smallest deviation among the values compared, an observation that should be read as indicative rather than as a demonstration of superior accuracy, since no formal uncertainty budget is available for the present measurement. These results provide a methodological basis for further studies

of pressure-broadening parameters and of the atmospheric applications of this transition.

Keywords

Ammonia (NH₃), Ultra-Stable Laser, Voigt Profile, Line-Shape Analysis, Frequency Metrology, Phase Locking

1. Introduction

Ammonia (NH₃) is one of the most extensively studied polyatomic molecules in high-resolution infrared spectroscopy. It plays a central role in interstellar and planetary astrophysics, where it is routinely used as a probe of the physical conditions of the interstellar medium, of the atmospheres of the giant planets, and of cometary comas [1]. On Earth, NH₃ is the dominant alkaline trace gas in the troposphere and the principal precursor of ammonium-containing fine particulate matter (PM_{2.5}); its emissions, dominated globally by agricultural activities such as livestock manure management and nitrogen fertilizer use [2] [3], are directly implicated in eutrophication, acidification of sensitive ecosystems, and indirect radiative effects through secondary aerosol formation [4] [5], with documented consequences for plant biodiversity and ecosystem resilience [3] [6]. Accurate knowledge of its rovibrational spectrum in the mid-infrared is therefore of interest both for fundamental molecular physics and for atmospheric remote sensing. NH₃ has a pyramidal equilibrium structure and a permanent dipole moment of 1.44 D [7]; its rotational transitions lie in the microwave domain, while its rovibrational transitions span the near- and mid-infrared [8]. A distinctive inversion motion, in which the nitrogen atom periodically tunnels through the plane of the three hydrogen atoms, splits every rovibrational level into symmetric (s) and antisymmetric (a) sub-levels and gives rise to the characteristic “saQ” notation used to label transitions such as the one investigated here [9] [10]. Several fundamental, overtone and combination bands of NH₃ have been characterized near 3 μm [9], and near 10 μm the ν₂ saQ(6, 3) line has been specifically exploited for precision metrology because of its intensity and its isolation within the 8 - 12 μm atmospheric window [11] [12].

The metrological interest of the ν₂ saQ(6, 3) transition dates back to its identification as a calibration standard by Urban *et al.* [13], from which an *ab initio* frequency of 28,953,695 MHz can be derived.

The present work addresses two complementary questions: i) how accurately and reproducibly can the ν₂ saQ(6, 3) transition of NH₃ be characterized using an ultra-stable laser operated under realistic laboratory conditions, and ii) to what extent does the choice of line-shape model, a simple Gaussian profile versus a more complete Voigt profile, affect the extraction of the spectroscopic parameters from a sizeable set of independently recorded spectra? To answer these questions, we

analyze 46 absorption spectra of the ν_2 saQ(6, 3) line recorded at 300 K and 10 Pa, and we compare the results and the numerical robustness of Gaussian and Voigt fits applied to the complete data set. The remainder of the paper is organized as follows. Section 2 reviews the line-broadening mechanisms relevant to the interpretation of the observed profiles. Section 3 describes the laser source and spectrometer architecture. Section 4 presents the data set and the automated fitting procedure. Section 5 details the two line-shape models used in the analysis. Section 6 reports the fit results, the residual analysis, and the comparison with the literature. Section 7 discusses the physical and practical implications of these results, and Section 8 places the work in the broader context of mid-infrared spectroscopy and its atmospheric applications, before concluding in Section 9.

2. Line-Broadening Mechanisms in Molecular Absorption Spectroscopy

The interpretation of the experimental profiles reported in Section 5 relies on a small number of well-established physical mechanisms that determine the shape of a molecular absorption line. This section summarizes these mechanisms and the corresponding mathematical description, which forms the theoretical basis for the Gaussian and Voigt models introduced in Section 5, and for the discussion of residual structure in Section 6.

2.1. Beer-Lambert Absorption Law

For a laser beam of initial intensity I_0 propagating through an absorbing gas, the transmitted intensity $I(\omega)$ decreases with propagation length z according to the differential absorption law $dI = -k(\omega)I(\omega)dz = -n\sigma(\omega)I(\omega)dz$, where $k(\omega)$ is the absorption coefficient, $\sigma(\omega)$ the absorption cross-section and n the molecular number density. Integration over a path length L yields the familiar Beer-Lambert law, $I(\omega) = I_0 \cdot \exp[-k(\omega)L] = I_0 \cdot \exp[-\alpha(\omega)]$, where $\alpha(\omega)$ is the absorption coefficient. For an ideal gas, it can be written by

$\alpha(\omega) = nLS_0\varphi(\omega - \omega_0) = (P/k_B T) \cdot LS_0\varphi(\omega - \omega_0)$, where S_0 is the line strength, P is the pressure, T is the temperature, and $\varphi(\omega - \omega_0)$ is the normalized line-shape function [8]. This function φ , is what the Gaussian and Voigt models of Section 5 are designed to reproduce.

2.2. Homogeneous and Inhomogeneous Broadening

The width of an absorption line results from several distinct mechanisms, conventionally separated into homogeneous mechanisms, which affect every absorbing molecule identically, and inhomogeneous mechanisms, which depend on individual molecular properties such as velocity. **Table 1** summarizes the mechanisms most relevant to the present study. Natural broadening, associated with the finite radiative lifetime of the excited state, produces a Lorentzian contribution that is typically below 100 Hz for the transitions considered here and is therefore negligible compared with the other mechanisms. Doppler broadening arises from the thermal (Maxwell-

Boltzmann) distribution of molecular velocities: each molecule absorbs at a frequency shifted according to its velocity component along the laser propagation direction, which produces an inhomogeneous, Gaussian line shape of half-width at half-maximum $\Delta\omega_D = (\omega_0/c)\sqrt{(2k_B T \ln 2/m)}$, where m is the molecular mass. Collisional (pressure) broadening results from the interruption of the coherent absorption process by intermolecular collisions; it produces a homogeneous, Lorentzian contribution of half-width $\Delta\omega_L = 2\pi B_p P$, proportional to the pressure P , with a broadening coefficient B_p typically of order 100 kHz/Pa for NH_3 [8]. Saturation broadening, a non-linear effect associated with high laser intensity, was not a limiting factor under the low-power conditions of the present measurements.

Table 1. Line-broadening mechanisms relevant to the interpretation of the NH_3 absorption profiles analyzed in this work.

Broadening Mechanism	Physical Origin	Line Shape	Typical Dependence
Natural	Finite radiative lifetime of the excited state	Lorentzian	Independent of pressure and temperature
Doppler	Thermal (Maxwell-Boltzmann) velocity distribution	Gaussian	$\propto T^{1/2}$ (T = temperature)
Collisional (Pressure)	Interruption of the absorption process by molecular collisions	Lorentzian	$\propto P$ (P = pressure)
Saturation (Power)	Non-linear response of the transition at high laser power	Modified homogeneous width	Depends on incident intensity
Dicke Narrowing	Velocity-changing collisions in a confined or dense medium	Narrowed Voigt-like (Galatry/Rautian)	$\propto P$, opposes Doppler broadening

2.3. The Voigt Profile and Beyond

When Doppler and collisional broadening are simultaneously significant, as is generally the case under the pressure and temperature conditions of the present measurements (10 Pa, 300 K), the resulting line shape is described by the Voigt profile, obtained from the convolution of the Gaussian and Lorentzian contributions (Equation (2), Section 5.2); the Voigt profile reduces to a pure Gaussian or a pure Lorentzian in the limits where one mechanism dominates the other. In sufficiently dense or confined gases, however, velocity-changing collisions can correlate the position and velocity of an absorbing molecule over the time scale of the measurement, producing a narrowing of the Doppler contribution known as the Dicke effect [14]. Two extensions of the Voigt profile have been developed to account for this narrowing: the Galatry (“soft-collision”) model, appropriate when the velocity of a heavy absorber is only weakly perturbed by collisions with a light perturber, and the

Rautian (“hard-collision”) model, appropriate when each collision fully randomizes the molecular velocity [15]. Both models introduce a narrowing parameter β , proportional to the perturber pressure, and both converge to the ordinary Voigt profile in the limit $\beta \rightarrow 0$. These advanced models are not fitted quantitatively in the present study, which is restricted to the Gaussian and Voigt profiles already available from the analysis of the 46-spectrum data set; they are introduced here because they provide the natural physical framework for interpreting the residual wing structure discussed in Section 6, and because they define a concrete direction for future refinements of the fitting procedure (Section 7.6).

3. Experimental Setup

3.1. Quantum Cascade Laser Operating Principle

A quantum cascade laser consists of a periodic stack of ultra-thin semiconductor layers of alternating composition, engineered by high-precision epitaxy so that the thickness of each layer controls, at the atomic scale, the energy levels and wave functions of the resulting quantum wells and barriers. Unlike conventional diode lasers, whose emission results from electron-hole recombination across the material bandgap, a QCL relies solely on the dynamics of electrons undergoing intersubband transitions within the conduction band [16]. Under an applied electric field, electrons tunnel sequentially through a large number of identical active regions, emitting one photon at each stage before being re-injected into the following period; each electron can therefore ideally generate as many photons as there are periods in the structure, giving QCLs their characteristic high output power for a compact device. This design flexibility allows QCLs to cover a spectral range extending from the mid-infrared to the terahertz domain (approximately 2.5 - 160 μm), with optimal performance typically obtained between 4 and 10 μm [16]. DFB QCLs, as used in the present spectrometer, combine a narrow intrinsic linewidth (often below 1 MHz) with a limited continuous tuning range of a few wavenumbers, in contrast to external-cavity QCLs, which trade some spectral purity for a much wider tunability; sub-kilohertz linewidths have in fact been demonstrated for free-running DFB QCLs referenced to a molecular sub-Doppler transition, illustrating the additional narrowing achievable without a full phase-locked architecture such as the one used here [17].

3.2. Laser Source and Spectrometer Architecture

The measurements analyzed in this work were carried out with a high-resolution mid-infrared spectrometer built around a distributed-feedback (DFB) quantum cascade laser (QCL), mounted in a Laboratory Laser Housing (LLH) equipped with a thermoelectric (Peltier) temperature controller. Operated at its set-point temperature, the QCL emits near 10 μm in continuous-wave, single-mode regime, and its emission frequency can be tuned across the R(6) line of the CO₂ laser used as the primary optical reference of the system [11].

The spectrometer, developed at the Laboratoire de Physique des Lasers (LPL), integrates a commercial DFB QCL, a free-space optical bench designed to minimize

parasitic reflections and interference, an electro-optic modulator (EOM), optical isolators, and high-sensitivity mid-infrared detectors. This architecture allows the continuous, reproducible acquisition of NH_3 absorption spectra with a frequency scale that is directly referenced to an ultra-stable optical standard, which is the central requirement for the high-resolution measurements reported here.

3.3. Frequency Stabilization and Metrological Traceability

Reaching the resolution and accuracy required for a metrological characterization of the transition requires stabilizing the QCL frequency against a well-defined reference. In the present setup, the QCL is phase-locked to a CO_2 laser that is itself frequency-stabilized on a saturated-absorption line of OsO_4 in a Fabry-Pérot cavity. This secondary reference is connected to the atomic clock ensemble of the LNE-SYRTE through an optical frequency comb operating near $1.5\ \mu\text{m}$, generated by a distributed-feedback diode and mixed in a non-linear AgAs_2 crystal; the resulting beat note between the CO_2 laser and the comb is detected, amplified, filtered, and measured with a frequency counter, closing the traceability chain to the international frequency standards.

The phase-lock loop itself compares the phase of the beat note between the QCL and the CO_2 laser, brought into the radiofrequency domain, with that of a reference synthesizer, and converts the resulting phase error into a correction voltage applied to the QCL through a proportional-integrator servo with a sign-inverting output stage. This scheme offers a wide correction bandwidth, requires no frequency modulation of either laser to generate the error signal, and preserves the tunability of the QCL. Under phase lock, more than 99.9% of the radiofrequency power of the beat note is concentrated in the coherent carrier, and the resulting QCL linewidth is reduced to the order of 10 Hz, three to four orders of magnitude narrower than in free-running operation [11]. To combine this narrow linewidth with a usable scanning range, the QCL is in practice locked to one of the two optical sidebands generated by the EOM, which are themselves tunable by several gigahertz through the microwave synthesizer driving the modulator; scanning the sideband frequency therefore allows the locked QCL to be tuned continuously over several gigahertz, more than eight orders of magnitude larger than its own linewidth, while retaining the stability transferred from the CO_2 reference.

Table 2 lists the successive components of this frequency-reference and traceability chain, from the primary time-and-frequency standard to the NH_3 absorption signal, together with the role of each component.

Table 2. Successive components of the frequency-reference and traceability chain linking the LNE-SYRTE atomic clock ensemble to the NH_3 absorption signal analyzed in this work.

Component	Role in the Traceability Chain
LNE-SYRTE atomic clock ensemble	Primary reference for time and frequency, traceable to the SI second
Optical frequency comb ($\approx 1.5\ \mu\text{m}$, RIO diode)	Links the atomic clock reference to the optical domain

Continued

Non-linear AgAs ₂ crystal	Mixes the comb output with the CO ₂ -laser beat signal
CO ₂ laser stabilized on OsO ₄	Secondary optical frequency reference, phase-compared to the comb
Phase-lock servo (proportional-integrator, sign-inverting stage)	Transfers the CO ₂ -laser stability to the QCL
DFB quantum cascade laser	Widely tunable spectroscopy source, phase-locked to the CO ₂ reference
Electro-optic modulator/YIG oscillator (6 - 18 GHz)	Generates the frequency-scanned optical sideband used for spectroscopy
Thermostated NH ₃ absorption cell	Sample under study, at controlled temperature and pressure
MCT detector (10 μ m)	Records the transmitted intensity as a function of scanned frequency

3.4. Absorption Cell, Frequency Scanning and Detection

For the measurements reported here, the frequency scan is performed by sweeping the microwave synthesizer that drives the EOM (YIG oscillator, 6 - 18 GHz), which shifts the frequency of the optical sideband on which the QCL is locked. The resulting beam is sent through an absorption cell containing NH₃ gas, held in a thermostated enclosure to control the sample temperature. The transmitted intensity is recorded with a mercury-cadmium-telluride (MCT) photodetector sensitive near 10 μ m. **Table 3** summarizes the resulting experimental conditions under which the 46 spectra analyzed in this work were recorded.

Table 3. Summary of the experimental conditions used to record and process the 46 NH₃ absorption spectra analyzed in this work.

Parameter	Value/Description
Target transition	ν_2 saQ(6, 3), ammonia (NH ₃)
Spectral region	Mid-infrared, $\approx 10 \mu\text{m}$ ($\approx 970 \text{ cm}^{-1}$)
Laser source	Distributed-feedback (DFB) quantum cascade laser, continuous-wave, single-mode
Frequency reference	CO ₂ laser stabilized on a saturated-absorption line of OsO ₄
Stabilization scheme	Optical phase-locked loop (proportional-integrator servo) between the QCL and the CO ₂ laser
Frequency scanning	Electro-optic modulator (EOM) driven by a YIG oscillator (6 - 18 GHz)
Absorption cell	Thermostated cell containing NH ₃ gas
Detector	MCT photodetector, 10 μm
Sample temperature	300 K
Sample pressure	10 Pa

Continued

Number of spectra recorded	46
Number of spectra with a robust Voigt fit	22 (of 46)
Fitting software	Python nonlinear least-squares routine (up to 200 iterations)

4. Data Set and Automated Processing**4.1. The 46-Spectrum Data Set**

A total of 46 absorption spectra of the ν_2 saQ(6, 3) transition of NH_3 were recorded under nominally identical experimental conditions, at a pressure of 10 Pa and a temperature of 300 K. The acquisition of a data set of this size, under fixed thermodynamic conditions, provides more than a single illustrative measurement: it offers a direct assessment of the internal consistency of the spectroscopic measurement chain. All 46 acquisitions display the same overall line shape: a broad, well-isolated absorption dip centered on the transition frequency, while the baseline level, the noise floor and, to a lesser extent, the apparent line width vary somewhat from one scan to the next. This variability, rather than being a shortcoming, is precisely the information that is needed to evaluate the robustness of an automated fitting procedure under realistic experimental conditions, and it is exploited throughout Sections 4.2 and 6 to distinguish genuine spectroscopic signal from acquisition-to-acquisition scatter.

4.2. Automated Fitting Procedure and Its Robustness

All 46 spectra were processed with the same automated, non-linear least-squares fitting procedure, implemented in Python using a nonlinear least-squares fitting routine, with up to 200 iterations. For every spectrum, the frequency of the point of maximum transmitted intensity was first identified and used to define an initial estimate of the line center; the data were then re-expressed relative to this initial estimate, and the fit was performed on the centered data set before the absolute center frequency was reconstructed by adding back the initial offset. Initial values of the remaining parameters: offset, amplitude, and characteristic width, were likewise determined automatically from the extrema of each spectrum, without manual adjustment. Applying an identical, non-interactive procedure to the full data set removes operator-dependent variability from the analysis and allows the 46 acquisitions to be compared on a strictly equal footing.

This uniform treatment also makes it possible to compare directly the numerical robustness of the two line-shape models introduced in Section 5. As shown in Section 6, the Gaussian model converged for all 46 spectra, whereas the Voigt model, which contains one additional free parameter and is more sensitive to the signal-to-noise ratio in the wings of the line, converged to a physically meaningful result for only 22 of the 46 acquisitions. This difference in convergence behavior

is itself a result of the study: it shows that the choice of line-shape model must balance physical completeness against the practical requirement of a stable, spectrum-by-spectrum fitting procedure when a sizeable and heterogeneous data set is processed automatically. It also indicates that a systematic quality pre-screening of the spectra (e.g., based on the local signal-to-noise ratio in the wings) could increase the effective Voigt convergence rate in future studies, without requiring any change to the underlying instrument.

4.3. Software Implementation and Parameter Initialization

The complete fitting pipeline: from the raw, frequency-referenced spectrum to the tabulated fit parameters, the overlaid fit curve, and the residual plot, was implemented as a single automated Python routine applied identically to each of the 46 spectra. For the Gaussian model, the routine first locates the frequency of maximum of the transmitted intensity to define the initial center estimate, recenters the data, and then sets the remaining initial parameters from the extrema of the recentered spectrum: the initial offset is set to the maximum observed, the initial amplitude to the difference between the maximum and the minimum, and the initial Gaussian width to a fixed default value of $\sigma_{\text{init}} = 10^7 \text{ Hz}$, appropriate for the frequency scale of the scan. For the Voigt model, the initial center is set to the mean frequency of the recorded points, the initial Gaussian and Lorentzian widths (σ_0, γ_0) are estimated from the effective width of the spectrum, the initial amplitude is set to $-(\text{max} - \text{min})$ of the recorded signal, and the initial baseline is set to the mean of the recorded signal. **Table 4** summarizes this initialization scheme, which is applied without manual adjustment to every spectrum in the data set, consistent with the fully automated character of the processing pipeline described in Section 4.2.

Table 4. Automatic initialization scheme used by the fitting pipeline for the Gaussian and Voigt models, applied identically to all 46 spectra.

Parameter	Gaussian Model (Equation (1))	Voigt Model (Equation (2))
Center (ω_0)	Frequency of maximum	Mean frequency of the recorded spectrum
Width	$\sigma_{\text{init}} = 10^7 \text{ Hz}$	σ_0, γ_0 estimated from the effective spectral width
Amplitude (A)	offset – minimum	$-(\text{maximum} - \text{minimum})$
Baseline (Offset/B)	Maximum	Mean of the recorded signal
Solver	Python nonlinear least-squares routine, ≤ 200 iterations	Python nonlinear least-squares routine, ≤ 200 iterations

5. Line-Shape Models

5.1. Gaussian Profile

In a dilute gas at thermal equilibrium, the random thermal motion of the absorbing molecules produces a Doppler shift of the absorption frequency that is distrib-

uted according to the Maxwell-Boltzmann velocity distribution. This inhomogeneous broadening mechanism gives rise to a Gaussian absorption line shape (Section 2.2). In the present analysis, the experimental spectra were first fitted with the Gaussian model

$$G(\omega) = \text{offset} - A \cdot \exp \left[-\frac{(\omega - \omega_0)^2}{2\sigma} \right] \quad (1)$$

where ω_0 is the central frequency of the transition, σ the Gaussian (Doppler) width, A the absorption amplitude, and offset the baseline level of the recorded signal. This model accounts for Doppler broadening only and neglects any contribution from molecular collisions.

5.2. Voigt Profile

At a finite pressure, collisions between NH_3 molecules interrupt the coherent absorption process and add a homogeneous, Lorentzian contribution to the line shape (Section 2.2). The resulting profile, obtained from the convolution of the Gaussian (Doppler) and Lorentzian (collisional) contributions, is the Voigt profile, expressed here as the real part of the Faddeeva function $w(z)$:

$$V(\omega) = A \cdot \text{Re} \left[w \left(\frac{\omega - \omega_0 + i\gamma}{\sigma\sqrt{2}} \right) \right] + B \quad (2)$$

where ω_0 is again the central frequency, σ the Gaussian width associated with Doppler broadening, γ the Lorentzian half-width associated with collisional broadening, A the amplitude, and B the baseline. For each spectrum, the initial values of ω_0 , σ and γ were estimated automatically from the mean position and effective width of the recorded profile, and A and B from its extrema, before the non-linear fit was performed. As discussed in Section 2.3, the Voigt profile is itself an approximation, valid in the absence of velocity-changing (Dicke-narrowing) collisions; it provides, in principle, a more complete physical description of the line shape than the Gaussian model, at the cost of an additional free parameter and, as discussed below, of a markedly reduced numerical robustness when applied indiscriminately to a heterogeneous set of experimental spectra.

The Voigt profile of Equation (2) can equivalently be written in its original integral form, as the convolution of a Gaussian and a Lorentzian line-shape function:

$$\Phi_V(\omega - \omega_0, \Delta\omega_D, \Delta\omega_L) = \frac{\sqrt{\ln 2}}{\Delta\omega_D} \sqrt{\pi} \cdot \left(\frac{a}{\pi} \right) \cdot \int_{-\infty}^{+\infty} \frac{e^{-y^2}}{(x - y)^2 + a^2} dy \quad (3)$$

with $a = \sqrt{2} \cdot (\Delta\omega_L / \Delta\omega_D)$ and $x = \sqrt{\ln 2} \cdot (\omega - \omega_0) / \Delta\omega_D$. In this notation, the parameter a quantifies the relative weight of the collisional (Lorentzian) and Doppler (Gaussian) contributions to the total line width: in the limit $a \rightarrow 0$, Equation (3) reduces to a pure Gaussian profile (Equation (1)), corresponding to the low-pressure limit in which Doppler broadening dominates; in the opposite limit of large a , it reduces to a pure Lorentzian profile, corresponding to the high-pressure limit in which collisional broadening dominates. The Faddeeva-function form of

Equation (2) used for the numerical fits in this work is mathematically equivalent to Equation (3), and is preferred for computation because efficient, high-precision numerical implementations of the Faddeeva function are widely available. This integral representation makes explicit the connection, discussed in Section 2.3, between the ordinary Voigt profile and the Galatry and Rautian profiles, which modify the same convolution integral to account for velocity-changing (Dicke-narrowing) collisions through an additional parameter β .

6. Results

6.1. Gaussian Fit: Central Frequency and Residuals

Figure 1 shows a representative spectrum (spectrum 1 of the data set) together with its Gaussian fit. The model reproduces the overall shape of the absorption feature, and in particular the central region of the line, reasonably well over the full 46-spectrum data set; the fitting procedure converged for every spectrum without manual intervention, confirming the numerical robustness of the Gaussian model under the present experimental conditions.

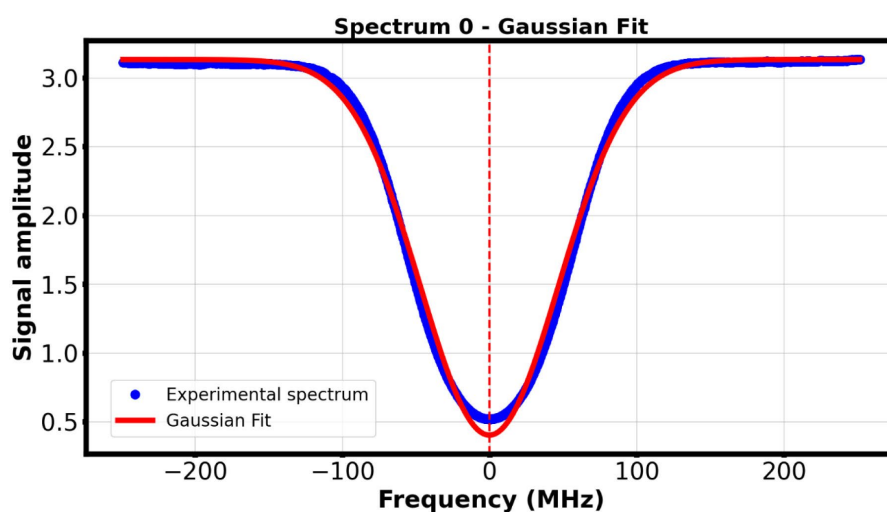


Figure 1. Representative NH_3 absorption spectrum (blue) and its Gaussian fit (red) as a function of frequency relative to the line center.

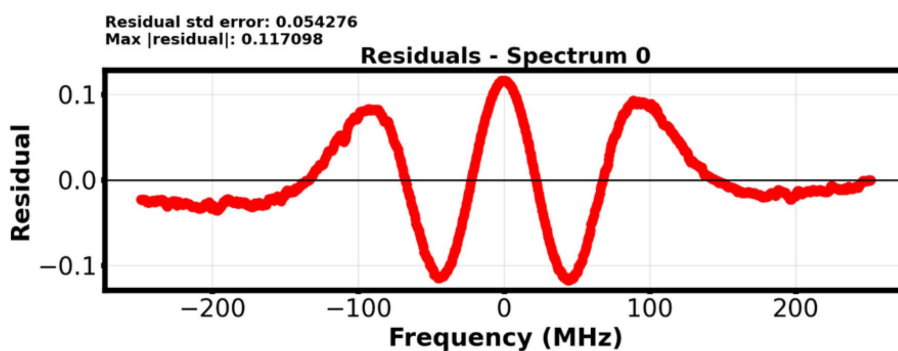


Figure 2. Residuals of the Gaussian fit shown in **Figure 1**, plotted as a function of frequency relative to the line center.

The residuals associated with this fit (**Figure 2**) reveal, however, a clear systematic structure rather than a random, noise-like pattern: the residual standard deviation is $\sigma(\text{residuals}) = 0.054$, with a maximum absolute residual of 0.117, and the residuals display a characteristic oscillatory pattern that is most pronounced near the line center and in the near wings. This behavior is the expected signature of a purely Doppler (Gaussian) model applied to a line that also contains a non-negligible collisional (Lorentzian) contribution: the discrepancy is systematic rather than statistical, and it is observed consistently across the data set.

6.2. Voigt Fit: Convergence and Residuals

Figure 3 shows the same spectrum fitted with the Voigt profile of Equation (2). When the fit converges, the model captures the central part of the line at least as well as the Gaussian model and is, by construction, better suited to represent the combined Doppler and collisional broadening expected under the experimental conditions of this study (10 Pa, 300 K). Applied to the complete data set, however, the Voigt fitting procedure converged to a stable, physically meaningful result for only 22 of the 46 recorded spectra. In the remaining cases, the fit either failed to converge or returned parameter values that were not physically acceptable (e.g., a negative or unrealistically large Lorentzian width), a behavior attributable to the variable signal-to-noise ratio across the data set, to residual baseline instabilities, and to the reduced spectral information available in the wings of lower-quality scans, where the additional Lorentzian parameter is least well constrained.

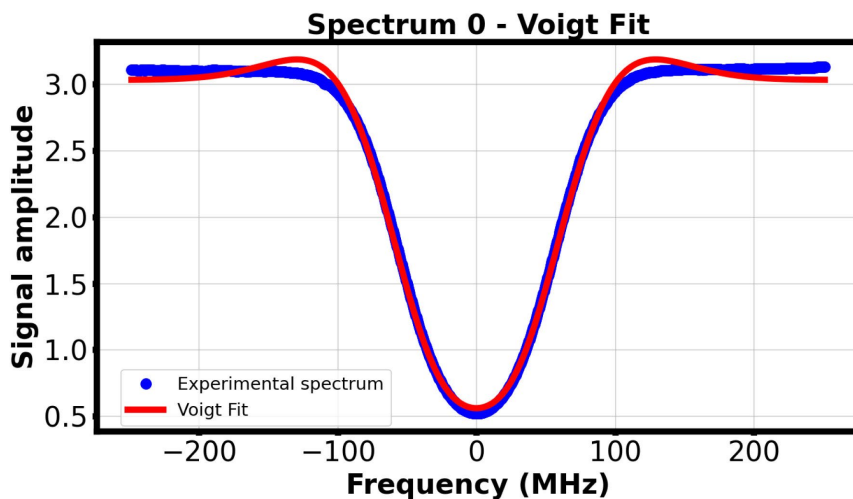


Figure 3. Representative NH_3 absorption spectrum (blue) and its Voigt fit (red) as a function of frequency.

Where the Voigt fit converges, the associated residuals are generally less systematic near the line center than those of the Gaussian model, consistent with the more complete physical content of the Voigt profile. Persistent oscillatory structure nonetheless remains visible in the far wings for several spectra, which we attribute to the combination of limited spectral resolution and reduced signal-to-

noise ratio in those regions (Section 2.3), rather than to an inadequacy of the Voigt model itself. The corresponding Voigt-fit residuals are shown in **Figure 4**.

6.3. Robustness of the Two Models: A Joint Result

Considered together, the Gaussian and Voigt analyses of the 46-spectrum data set constitute a methodological result in their own right, independent of the specific frequency value obtained. The Gaussian model is numerically robust; it converges systematically and yields a stable estimate of the line position across the full data set, but it is physically incomplete, as confirmed by the structured residuals of **Figure 4**. The Voigt model is physically more appropriate, but its successful application depends critically on the quality of the individual spectrum, so that only about half of the acquisitions (22 of 46) yield an exploitable Voigt fit. This contrast should not be interpreted as a rejection of collisional broadening in the present system; rather, it reflects a practical trade-off, under the present experimental conditions, between the physical completeness of a line-shape model and the numerical stability required to apply it uniformly to a realistic, heterogeneous set of experimental spectra.

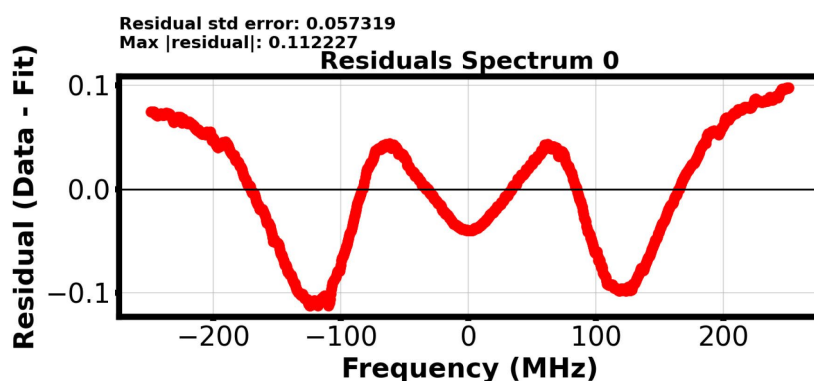


Figure 4. Residuals of the Voigt fit shown in **Figure 3**, plotted as a function of frequency relative to the line center.

6.4. Baseline and Amplitude Behavior of the Two Models

A further qualitative difference between the two models is visible by comparing **Figures 2-4**. The fitted baseline level in the Gaussian fit tracks the far wings of the spectrum closely, whereas the Voigt fit, whose slowly decaying Lorentzian component extends further from the line center, tends to settle on a marginally different effective baseline over the same frequency window. This behavior is consistent with the functional difference between the two profiles: a Gaussian decays much faster than a Lorentzian away from the line center, and it illustrates, at the level of the baseline and amplitude parameters rather than of the center frequency, the same physical trade-off already discussed for the residuals in Sections 6.1 - 6.3: a model that includes the Lorentzian component redistributes part of the fitted intensity between the amplitude, the baseline and the width parameters, which further underlines the importance of reporting the complete set of fitted parameters,

and not only the central frequency, whenever the line-shape model is changed.

6.5. Comparison with Previous Frequency Determinations

Table 5 compares the central frequency of the ν_2 saQ(6, 3) transition obtained in the present work with the ab initio value of Urban *et al.* [13] and with three previous experimental determinations: two obtained by CO₂-laser-based saturated-absorption or sideband techniques [18] [19], and one obtained with an earlier phase-locked QCL spectrometer of the same design lineage [11].

Table 5. Comparison of the absolute frequency of the ν_2 saQ(6, 3) transition of NH₃ reported in the literature with the value obtained in this work. Deviations are expressed relative to the ab initio value of Urban *et al.* [13], taken as a common reference (0 kHz); figures in parentheses are the 1 σ uncertainties reported by the original authors.

Source	Method	Frequency (MHz)	Deviation from ab Initio (kHz)
Urban <i>et al.</i> (1983) [13]	Ab initio calculation	28,953,695 (reference)	0
Lemarchand <i>et al.</i> (2011) [18]	CO ₂ -laser saturated-absorption spectroscopy	28,953,693.9(1)	−1100.0
Magerl <i>et al.</i> (1984) [19]	CO ₂ -laser microwave-sideband saturation spectroscopy	28,953,694.010(70)	−990.0
Sow (2015) [11]	Phase-locked QCL spectrometer	28,953,694.001(20)	−999.0
This Work	Phase-locked QCL spectrometer, Gaussian/Voigt fit	28,953,694.0613(47)	−938.712

All four experimental determinations lie within about 200 kHz of one another and within roughly 1 MHz of the ab initio prediction, which is consistent with the expected accuracy of high-level ab initio rovibrational calculations for a molecule as light and as strongly coupled as NH₃. The frequency obtained in the present work, 28,953,694.06 MHz, differs from the ab initio value by −938.712 kHz, which is numerically the smallest deviation among the four values compared, smaller by about 88 kHz than the closest previous experimental determination [19] and by about 97 kHz than the QCL-based measurement of Sow [11]. We emphasize, however, that this comparison should be read with caution: unlike the three literature values, the present measurement is not accompanied by a formal budget of statistical and systematic uncertainties (e.g., residual pressure shift, distribution of fitted centers across the 46 spectra, or servo-loop residual noise), so that the closer numerical agreement with the ab initio value is best regarded as indicative of the overall consistency of the measurement with previous work, rather than as evidence of a higher intrinsic accuracy. **Figure 5** illustrates this comparison graphically.

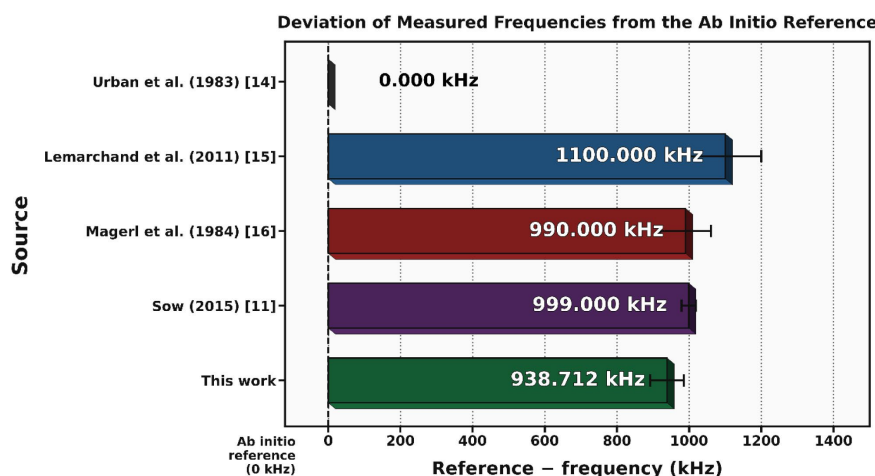


Figure 5. Deviation, in kHz, of four determinations of the frequency of the ν_2 saQ(6, 3) transition of NH_3 from the ab initio value of Urban *et al.* [13], taken as the reference (0 kHz).

6.6. Reproducibility of the Automated Fitting Procedure across the 46 Spectra

The results presented above are illustrated using a representative spectrum, but the same automated fitting procedure was applied independently and without manual adjustment to all 46 spectra recorded under nominally identical experimental conditions. The Gaussian model converged for all 46 acquisitions, whereas physically acceptable Voigt solutions were obtained for 22 spectra. This systematic application of an identical processing procedure demonstrates the numerical reproducibility and robustness of the Gaussian fitting approach across the complete experimental data set, while also highlighting the greater sensitivity of the Voigt model to variations in spectral quality.

The 46/46 Gaussian convergence rate should not, however, be interpreted as a quantitative measure of the statistical uncertainty of the reported central frequency. Such an assessment would require the individual fitted center frequencies and their dispersion, which are not reported in the present analysis. The present result therefore establishes the robustness of the automated fitting procedure rather than a complete statistical uncertainty budget for the transition frequency.

6.7. Framework for Extracting Thermodynamic Parameters

Beyond the central frequency, the width parameters returned by the Gaussian and Voigt fits are, in principle, directly connected to the thermodynamic conditions of the absorbing sample. Inverting the Doppler-width expression of Section 2.2 gives $T = (mc^2/2k_B) \cdot (\Delta\omega_D/\omega_0)^2$, so that a measurement of the Doppler width $\Delta\omega_D$ (the Gaussian σ parameter of Eq. 1) directly yields the gas temperature once the molecular mass m and the fixed constants c and k_B are specified. Similarly, the Lorentzian half-width γ returned by the Voigt fit (Equation (2)) is expected to scale linearly with pressure, $\gamma \propto P$, providing a route to a pressure-broadening coefficient for the ν_2 saQ(6, 3) transition. In the present study, these relations are

reported as the methodological framework that connects the fitted line-shape parameters to physical observables; a quantitative determination of temperature or of a pressure-broadening coefficient from the width parameters of the 46 fitted spectra was not carried out here and is identified in Section 8 as a direct extension of the present analysis, to be pursued once a dedicated uncertainty budget for the fitted widths is established.

7. Discussion

7.1. Why the Gaussian Fit Remains a Useful Practical Tool

It might appear inconsistent to conclude, on physical grounds, that the Voigt profile provides the more appropriate description of the line shape, and yet to rely in practice on the Gaussian model for a robust, data set-wide estimate of the line position. These two statements are not in conflict. The choice of the Gaussian model as the primary tool for extracting the central frequency across the full 46-spectrum data set should not be interpreted as a rejection of collisional broadening in the sample; it reflects instead a practical trade-off associated with the quality and resolution of the available spectra. Under the pressure and temperature conditions of this study (10 Pa, 300 K), the Lorentzian contribution to the line shape, while measurable when the Voigt fit converges, is small enough that the Gaussian model already captures the dominant part of the absorption feature and, in particular, the position of its center. The systematic residual structure discussed in Section 6.1 shows that this simplification is not free of physical cost, but it does not compromise the primary quantity of interest, the central frequency, to an extent that is inconsistent with the level of agreement observed with the literature values in **Table 5**.

7.2. Residual Structure and Its Physical Interpretation

The analysis of the fit residuals is not a secondary check but a source of information in its own right about the adequacy of each line-shape model. For the Gaussian model, the systematic, oscillatory residual pattern observed for essentially every spectrum in the data set (illustrated in **Figure 2**) indicates that a purely Doppler-broadened profile cannot fully represent the experimental line shape, even though the fitted center frequency remains well defined. This is the expected behavior when a non-negligible collisional contribution is present but is not included in the model. For the Voigt model, the residuals are generally less structured near the line center, in agreement with the more complete physical content of Equation (2), but persistent oscillations remain visible in the wings for several spectra. As discussed in Section 2.3, part of this residual wing structure may also reflect Dicke narrowing, which a plain Voigt profile does not capture; we attribute the observed residual wing structures primarily to the reduced signal-to-noise ratio and to the limited spectral resolution in the wings of the absorption feature, rather than to a fundamental inadequacy of the Voigt profile itself, but a quantitative separation of these two contributions would require fitting the Galatry or

Rautian models directly, which was not undertaken in the present study.

7.3. Value of the Phase-Locked Frequency-Traceability Chain

Beyond the specific comparison of line-shape models, the frequency-traceability chain described in Section 3.3 is itself a central feature of the present measurement. Phase-locking the QCL to a CO₂ laser stabilized on a saturated-absorption line of OsO₄, and connecting this secondary reference to the LNE-SYRTE atomic clock ensemble through an optical frequency comb, provides a well-defined and internally consistent frequency scale for every one of the 46 spectra analyzed in this work. This architecture combines the wide, continuous tunability required to scan across the ν_2 saQ(6, 3) line, obtained by locking the QCL to an electro-optically generated sideband, with a spectral purity and long-term stability that would not be accessible with a free-running QCL alone. It is this combination, rather than any single component, that makes the QCL spectrometer used here a suitable instrument for the kind of frequency-referenced, high-resolution measurement reported in this paper, and that would similarly benefit future studies of other rovibrational transitions of NH₃ or of other mid-infrared-active molecules.

7.4. Comparison with Previous QCL-Based and CO₂-Laser-Based Measurements

The two CO₂-laser-based determinations included in Table 5 [18] [19] rely on saturated-absorption or microwave-sideband saturation techniques that have been used for several decades and are well characterized, but whose frequency coverage around a given CO₂ laser line is comparatively narrow. The QCL-based determination of Sow [11], obtained with an earlier realization of a phase-locked QCL spectrometer developed at the same laboratory, shares the essential architecture used in the present work: phase locking to a stabilized CO₂ reference, and the two measurements should accordingly be regarded as complementary determinations obtained with closely related instrumentation, rather than as fully independent tests of the transition frequency. The present measurement nonetheless contributes an additional, independently processed data set (46 spectra, automated Gaussian and Voigt fitting) and confirms, within experimental scatter, the frequency reported in [11]. A fully independent test of the transition frequency would require an instrument based on a different reference chain (for example, a direct comb-referenced measurement without an intermediate CO₂ laser), which lies beyond the scope of the present study.

7.5. Three Levels of Contribution

The results of this study can be organized on three complementary levels. The first is experimental: the ν_2 saQ(6, 3) transition of NH₃ has been measured with a phase-locked QCL spectrometer, yielding a central frequency of 28,953,694.0613 MHz. The second is methodological: the automated, spectrum-by-spectrum processing of 46 independently recorded scans makes it possible to quantify, within this data

set, the relative robustness of Gaussian and Voigt fitting under realistic experimental conditions: full convergence for the Gaussian model against a 22-out-of-46 convergence rate for the Voigt model. The third is scientific: the resulting frequency is consistent with the previously reported experimental values and with the *ab initio* prediction, which confirms the identification of the transition and validates the overall performance of the phase-locked QCL architecture for high-resolution mid-infrared spectroscopy. Together, these three levels define the contribution of the present work, without requiring any additional calculation beyond those already performed on the recorded data set.

7.6. Towards More Advanced Line-Shape Models

The residual structure discussed in Sections 6 and 7.2, together with the reduced convergence rate of the Voigt fit, suggests that further improvements to the fitting procedure could be pursued in future work without requiring new instrumentation. Two directions appear particularly promising. First, fitting the Galatry or Rautian profiles introduced in Section 2.3 to the subset of high-quality spectra could quantify the contribution of Dicke narrowing to the residual wing structure and potentially improve the convergence rate relative to the plain Voigt model, since these profiles are specifically designed for the pressure regime explored here. Second, a systematic, quantitative quality metric for each spectrum (for example, a local signal-to-noise ratio computed automatically in the wings before fitting) could be used to pre-select the subset of spectra best suited to a Voigt or Galatry-Rautian analysis, rather than discovering the Voigt convergence outcome only after the fit is attempted, as was done in the present study.

7.7. Experimental Limitations

Several limitations of the present measurements should be stated explicitly. First, the quality of the 46 recorded spectra is not uniform: variations in signal-to-noise ratio and in baseline stability directly affect the convergence and the reliability of the Voigt fit, and are the primary reason why fewer than half of the acquisitions yield an exploitable Voigt fit. Second, instrumental effects, including residual amplitude modulation, imperfect baseline correction, and the finite spectral resolution in the wings of the line, are likely to contribute to the structured residuals observed with both models, and cannot be fully disentangled from genuine collisional-broadening or Dicke-narrowing effects on the basis of the present data set alone. Third, no formal budget of statistical and systematic uncertainty has been established for the central frequency reported here. Although the Gaussian fitting procedure converged successfully for all 46 spectra, corresponding to a convergence rate of 100%, this numerical robustness should not be interpreted as a quantitative measure of the statistical uncertainty of the reported frequency. Such an assessment would require, in particular, the individual fitted center frequencies and their dispersion, together with an evaluation of systematic contributions such as residual pressure and power shifts. These limitations do not affect the identifi-

cation of the transition or the overall consistency of the measurement, but they do restrict the strength of the metrological claims that can be drawn from the present data set.

7.8. On the Use of the Ab Initio Value as a Comparison Reference

Table 5 and **Figure 5** use the ab initio value of Urban *et al.* [13] as a common zero for comparing the four frequency determinations, but this choice deserves a methodological comment. Ab initio rovibrational calculations for a light, strongly anharmonic and Coriolis-coupled molecule such as NH_3 are not, in general, more accurate than the best available experimental determinations; the stated accuracy of the calibration standard used here (better than $1 \times 10^{-4} \text{ cm}^{-1}$, equivalent to about 3 MHz) is in fact coarser than the sub-100 kHz uncertainties reported for the CO_2 -laser-based and QCL-based experimental measurements in **Table 5**. The ab initio value therefore functions in this comparison as a convenient, independent common reference point rather than as a metrological standard of superior accuracy. Consequently, the fact that the present measurement lies numerically closest to this ab initio value (Section 6.5) should not be read as a claim that it is the most accurate determination of the transition frequency; a rigorous ranking of the experimental determinations by accuracy would instead require comparing their respective, independently established uncertainty budgets, which, as noted in Sections 6.5 and 7.7, is not available for the present measurement, and remains an open task for future work.

7.9. Reproducibility and Prospects for Independent Replication

Because the fitting pipeline described in Sections 4.2 and 4.3 is fully automated and requires no manual, spectrum-by-spectrum intervention, the analysis presented in this paper is, in principle, directly reproducible from the recorded frequency-referenced spectra, and could be extended straightforwardly to additional spectra recorded with the same or a similar instrument. This reproducibility is itself a methodological asset distinct from the specific frequency value reported: it means that the comparison between Gaussian and Voigt convergence rates established here (22 of 46 spectra for the Voigt model) can be used as a quantitative reference against which future improvements to the fitting procedure, for example, the Galatry- or Rautian-based analysis outlined in Section 7.6, or an improved quality pre-screening of the spectra, can be evaluated on the same data set or on newly recorded ones.

7.10. Relevance to the Boltzmann-Constant Determination Program

The ν_2 saQ(6, 3) transition of NH_3 has been used, in earlier work by the same research community, as the basis for a spectroscopic determination of the Boltzmann constant k_B from the Doppler width of the absorption profile, exploiting the relation $T = (mc^2/2k_B)(\Delta\omega_D/\omega_0)^2$ introduced in Section 6.5 at a fixed, in-

independently controlled temperature [11] [12] [20]. Such a determination places stringent requirements on the accuracy of the fitted Doppler width and on the control of pressure- and power-related systematic effects, well beyond what is required for a line-position measurement alone. The present results, in particular the quantified difference in convergence and residual behavior between the Gaussian and Voigt models (Sections 6.1 - 6.3), are directly relevant to that broader metrological program: they show that a purely Gaussian fit, while adequate for the central frequency, would need to be corrected for the systematic bias introduced by unmodeled collisional broadening before its width parameter could be used for a Boltzmann-constant-type measurement, reinforcing the case for the Voigt-based (or Galatry/Rautian-based) analysis discussed in Section 7.6 whenever the line width itself, rather than only its center, is the quantity of interest.

8. Scientific Significance and Perspectives

8.1. Broader Significance for Mid-Infrared Spectroscopy

Beyond the specific transition investigated here, the results demonstrate that the system can be effectively employed for the high-resolution characterization of rovibrational transitions of NH_3 , and by extension of other mid-infrared-active molecules, such as CH_4 , NO_2 , CO , or small volatile organic compounds. The methodological comparison between Gaussian and Voigt fitting, and the explicit quantification of the convergence rate of the more complete model, provide a practical guideline for future studies that must process large numbers of experimental spectra in an automated fashion: a simple, numerically robust model may be preferable for the systematic extraction of line positions across a heterogeneous data set, while a more complete model should be reserved for the subset of spectra whose quality supports it, or complemented by improved signal-to-noise ratio and baseline stabilization.

The results are also relevant to the maintenance and refinement of spectroscopic reference databases such as HITRAN and GEISA, which compile the line positions, intensities, and broadening parameters used throughout atmospheric radiative-transfer and remote-sensing models. An additional, independently obtained determination of the ν_2 saQ(6, 3) frequency, consistent with previous measurements within the precision discussed in Section 6.5, contributes to the cross-validation of the reference value used in such databases, even though, as stressed in Section 7.7, the absence of a formal uncertainty budget for the present measurement means that it should be regarded as a consistency check rather than as a candidate for direct database revision.

8.2. Atmospheric Perspective

The position and shape of NH_3 absorption lines depend on the thermodynamic conditions of the absorbing medium, through Doppler broadening (temperature) and collisional broadening and shift (pressure and composition); this dependence is, in principle, the physical basis for using rovibrational spectroscopy of NH_3 to

probe atmospheric conditions. The spectroscopic parameters characterized in this work: the central frequency of the ν_2 saQ(6, 3) transition, the framework of Section 6.5 relating the fitted widths to temperature and pressure, and the relative performance of Gaussian and Voigt models in fitting the observed profiles, constitute a methodological basis for such future developments. We stress, however, that the atmospheric application of this transition is not experimentally validated in the present study: no pressure- or temperature-dependent series of measurements was performed, and no atmospheric sample or field deployment was involved. This aspect should accordingly be regarded as a perspective for future work rather than as a demonstrated outcome of the present measurements. Realizing this perspective would in practice require coupling the present line-position measurement with the kind of aerosol-formation [21] and heterogeneous-landscape flux [22] parameterizations already used in atmospheric chemistry-transport and deposition models of NH_3 , for which accurate input line parameters are a recognized limiting factor.

8.3. Extension to Airborne and Ground-Based Remote Sensing

Phase-locked QCL spectrometers of the type used in this work are related, in their source technology, to the compact QCL-based instruments increasingly deployed for ground-based and airborne in-situ measurements of atmospheric trace gases, where high spectral selectivity and fast time response are required to resolve rapidly varying concentrations along a flight path or across a monitoring network [23] [24], including ultra-sensitive heterodyne detection schemes operating within the same atmospheric transmission windows [25]. Ground-based multi-gas microsensor networks developed for ambient air-quality surveillance [26] [27] illustrate a complementary, lower-cost class of instrumentation that could similarly benefit from an accurately referenced line position. While the laboratory-scale, frequency-comb-referenced architecture described in Section 3 is not itself designed for field deployment, the spectroscopic parameters it provides: an accurately referenced line position and a validated line-shape analysis procedure, are precisely the inputs required to calibrate and to interpret the simpler, non-comb-referenced QCL sensors used in such deployments. In this sense, the present laboratory characterization and field-oriented QCL sensing are complementary steps of the same measurement chain, from a validated spectroscopic parameter to an operational atmospheric measurement.

8.4. Further Directions

Beyond the specific improvements to the fitting procedure discussed in Section 7.6, the methodology validated in this work suggests several directions for future studies. The same automated processing chain could be applied to other rovibrational transitions of NH_3 , including those in the ν_1 and ν_3 bands, to build a more complete, internally consistent map of the lines most suitable for detection and metrology. The same phase-locked QCL architecture is, in principle, transferable

to other mid-infrared-active atmospheric species (CH_4 , NO_2 , CO , volatile organic compounds), which would allow a common, traceable frequency reference to be used across a multi-species spectroscopic data set. Finally, pursuing the pressure-broadening framework outlined in Section 6.5, together with the Galatry and Rautian models introduced in Section 2.3, would provide the quantitative link between the present line-position measurement and the parameters required for atmospheric radiative-transfer applications, and would allow the accuracy claims of Section 6.4 to be placed on a firmer, uncertainty-budgeted footing.

9. Conclusion

This study has characterized the ν_2 saQ(6, 3) rovibrational transition of ammonia experimentally using an ultra-stable laser. Forty-six absorption spectra were recorded at 300 K and 10 Pa and processed with an identical, automated non-linear fitting procedure. The Gaussian model provided a robust, uniformly convergent estimate of the transition frequency across the complete data set, while the Voigt model, though physically more complete since it accounts for both Doppler and collisional broadening, converged for only 22 of the 46 spectra, reflecting its greater sensitivity to the quality of the experimental data. The resulting central frequency, 28,953,694.0613 MHz, is consistent with previous experimental determinations reported in the literature and with the *ab initio* prediction, within a spread of about one megahertz that is typical of this transition. These results demonstrate the suitability of a phase-locked QCL architecture for high-resolution spectroscopy of NH_3 in the mid-infrared, and they provide a methodological basis, in particular regarding the trade-off between the physical completeness and the numerical robustness of line-shape models, and the framework linking fitted line widths to temperature and pressure, for future studies of pressure-broadening parameters of NH_3 and of its use as a spectroscopic probe in atmospheric applications.

Author Contributions

P.L.T.S. and F.D.: conceptualization, simulation, formal analysis, and writing—original draft. S.M.: supervision, project administration, validation, and writing—review. Y.G. and M.L.S.: methodology, formal analysis, and writing—review. All authors reviewed and approved the final manuscript.

Data Availability

The frequency-referenced spectra and the fitted parameters analyzed in this study are available from the corresponding author upon reasonable request.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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Nomenclature

The following symbols and abbreviations are used throughout this paper.

Symbol/Abbreviation	Meaning
ω_0, ν_0	Central (unperturbed) frequency of the transition
$\Delta\omega_D$	Doppler half-width at half-maximum
$\Delta\omega_L$	Collisional (Lorentzian) half-width at half-maximum
Σ	Gaussian width parameter (Doppler)
Γ	Lorentzian width parameter (collisional)
B	Dicke-narrowing (velocity-changing collision) parameter
A, B/A, offset	Fit amplitude and baseline parameters
S_0	Line strength
k_B	Boltzmann constant
M	Molecular mass of NH_3
P, T	Pressure and temperature of the absorbing sample
$\varphi(\omega - \omega_0)$	Normalized line-shape function
QCL	Quantum cascade laser
DFB	Distributed feedback (laser)
HITRAN/GEISA	Mid-infrared/atmospheric molecular absorption spectroscopic databases