The Ground and First Excited Torsional States of Acetic Acid

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A global fit of microwave and millimeter-wave rotational transitions in the ground and first excited torsional states ($v_t = 0$ and 1) of acetic acid (CH₃COOH) is reported, which combines older measurements from the literature with new measurements from Kharkov, Lille, and NIST. The fit uses a model developed initially for acetaldehyde and methanol-type internal rotor molecules. It requires 34 parameters to achieve a unitless weighted standard deviation of 0.84 for a total of 2518 data and includes A- and E-species transitions with $J \le 30$ and $K_a \le 15$. While these results represent a significant improvement over past fitting attempts, it should be cautioned that the present data set is dominated by $v_t = 0$ transitions, and no direct infrared measure of the $v_t = 1 \leftarrow 0$ torsional interval is available. © 2001 Academic Press

I. INTRODUCTION

Acetic acid (CH₃COOH) is of astrophysical interest, since several of its rotational transitions have recently been detected in space (1). There is considerable interest in the detection of interstellar acetic acid because in the laboratory a bimolecular synthesis of the simplest important amino acid, glycine (NH₂CH₂COOH), occurs when acetic acid combines with NH₂⁺, followed by release of a proton. Furthermore, the detection of isomeric pairs such as acetic acid and methyl formate HCOOCH₃ (which has been found with large column densities in Orion A and Sgr B2 (e.g., Refs. (2-4)) is important for the understanding of chemical reactions in the interstellar medium. In spite of intensive radio searches, however, and in spite of the relatively intense rotational spectrum expected from its dipole moment components $\mu_b = 1.47$ D and $\mu_a = 0.86$ D (5) $(1 D = 3.33564 \times 10^{-30} Cm)$, the first radio detection of acetic acid was only realized in 1997 in the Sgr B2 large-molecule Heimat source (1), using frequencies of the $8_{*,8}$ – $7_{*,7}$ A and E lines measured at 90 246.26(5) and 90 203.35(5) MHz, respectively, at the National Institute of Standards and Technology (NIST). The $9_{*,9}$ – $8_{*,8}$ A and E lines of CH₃COOH calculated at 100 897.83 and 100 855.02 MHz, respectively, by Wlodarczak and Demaison (6) were subsequently used to confirm this detection. Each of the lines with a * in place of K_a consists of four unresolved overlapping transitions, two a-type and two

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b-type. The 90 203-MHz line, for example, contains the a-type 8_{08} - 7_{07} E and 8_{18} - 7_{17} E transitions and the b-type 8_{08} - 7_{17} E and 8_{18} - 7_{07} E transitions (I).

A fair amount of spectroscopic ground state work is available in the literature for the acetic acid molecule (5-10). A very early study was published in 1957 by Tabor (7), who measured 11 lines for the normal species and 13 lines for the deuterated species. Later Krisher and Saegebarth measured and assigned 77 torsional ground state lines for the normal species and 54 lines belonging to the deuterated species CD₃COOH (5). Using the principal axis method (PAM) and a model extended to include perturbation terms through sixth order, they were able to determine a value for the torsional barrier of $V_3 = 168.2(2)$ cm⁻¹ (480.8(5) cal/mol). Stark-effect measurements gave electric dipole moment components of $\mu_a = 0.86(1)$ D and $\mu_b =$ 1.47(2) D. In 1981 van Eijck et al. (8) performed 67 new measurements and remeasurements in the frequency range from 27 to 40 GHz in the ground torsional state ($v_t = 0$) of acetic acid. Using both the PAM and IAM (internal axis method), they succeeded in fitting the A-species frequencies to experimental precision, but the E-species root-mean-square (rms) deviations were 1.64 and 0.70 MHz for the PAM and IAM methods, respectively. In 1982, Demaison et al. (9) measured lines in the 70-290 GHz and 28-39 GHz ranges, but again the rms deviations were large for E-species lines. The first excited torsional state ($v_t = 1$) of acetic acid was investigated by van Eijck and collaborators up to J = 21 in 1983 (10). The observed-minus-calculated values showed severe discrepancies, some as large as several megahertz. In 1988, Demaison and Wlodarczak (6) extended measurements of the ground torsional state to frequencies up to 362 GHz.



The present study reports a global fit of over 2500 rotational transitions in acetic acid, using the same rho axis method (RAM) that was previously applied to the four lowest torsional states of acetaldehyde CH3CHO (11) and the two lowest torsional states of methanol CH₃OH (12). Just as for those two molecules (13, 14), we want ultimately to provide a frequency and intensity atlas of measured and calculated lines that can be used for further astrophysical detection, but it seemed prudent as a first step to establish confidence limits and identify shortcomings in the theoretical model and experimental data set. Transition frequencies were taken from four sources (which for various reasons were put into the fit in the following chronological order): (i) most measurements published in the references listed above (provided they were not remeasured as part of the present work), (ii) new lines from Lille, measured using broadband recording in various frequency intervals in the region from 148 to 250 GHz, (iii) new lines from NIST, both molecular-beam-cooled Fourier transform lines from 8 to 40 GHz, and submillimeter lines from 78 to 118 GHz, 300 to 304 GHz, and 350 to 365 GHz, and (iv) published (15) (A-species) and unpublished (E-species) lines from Kharkov, in the region from 49 to 155 GHz. Our main effort was focused on the torsional ground state, but several hundred $v_t = 1$ transitions are also included in the present fit.

II. EXPERIMENTAL DETAILS

Measurements from Lille

Measurements in Lille were made in the 148–250 GHz range. The millimeter-wave sources were second and third harmonics of a 74–80 GHz phase-locked Gunn diode and a Russian Istok backward-wave oscillator working in the 170–250 GHz range. The lines were detected with a liquid-Hecooled InSb bolometer. The spectrometer was used in a scanning mode with a frequency step of 50 kHz. The uncertainty of the measurements is estimated to be less than 100 kHz. The sample of CH₃COOH was obtained from Aldrich Chemical Co., Inc. (16) and used without further purification.

Measurements from NIST

Fourier transform microwave (FTMW) measurements at NIST were carried out from 8 to 40 GHz. A sample consisting of 6.5 Torr (1 Torr = 133.3 Pa) of glacial acetic acid was placed in an evacuated cylinder. The tank was then pressurized to 5000 Torr with an inert carrier gas consisting of 20% helium in neon by volume, resulting in a mixture of 0.13% by volume. For measurements below 26.5 GHz, one of the standard NIST instruments was used. The electronics, hardware, and software are identical to the smaller FTMW instrument (17). For measurements between 26.5 and 40 GHz, some additional microwave hardware was used. Referring to Fig. 3 of Ref. (17), active doublers were inserted after the programmable attenuators and between S1 and the image rejection mixer. The mi-

crowave amplifier located immediately after the Fabry–Perot cavity, S3, and the image rejection mixer were replaced with units designed to operate in the 26.5–40 GHz region. The 60-MHz intermediate frequency (IF) signal from the mixer was mixed with a 90-MHz signal. The resulting difference frequency (30 MHz) was then digitized directly at 8 MHz as described in Ref. (17). In all of these experiments the pulsed nozzle was mounted on the back side of one of the Fabry–Perot cavity mirrors so that the gas pulse travels coaxially with the Fabry–Perot cavity axis (18). This results in lines with about 10 kHz full width at half-maximum. As described in Ref. (17), all frequencies are referenced to a 10-MHz frequency obtained from a Loran-C receiver. This frequency is referenced to a Cs-clock which has stability on the order of 1 part in 10¹¹.

For measurements in the 300–304 and 350–365 GHz spectral regions, a spectrometer consisting of a broadband tunable ISTOCK backward-wave oscillator, phase-locked to a precision-tunable 53–78 GHz KVARTZ synthesizer was used. The signal passes through an absorption cell 3 m long and is focused on a high-sensitivity liquid-He-cooled InSb hot-electron bolometer. For measurements in the 78–118 GHz range a broadband precision-tunable KVARTZ synthesizer was used directly as the source. In both cases the cell was maintained at 7 mTorr of acetic acid under mild flow conditions to avoid impurities. Frequency detection was the same as for the FTMW measurements.

Measurements from Kharkov

Measurements of the spectra were carried out using an automated spectrometer in the millimeter region (19). The spectrometer employs a computer-controlled synthesizer in the frequency range of 50-120 GHz as a source of millimeterwave radiation. It is designed for detailed investigation of the spectra of molecules with very high resolution. The synthesis of the frequencies in the millimeter region is carried out by a two-step frequency multiplication of the reference synthesizer (390-400 MHz) in two phase-lock-loop (PLL) stages. The first PLL stage utilizes a Klystron generator in the range of 3.4–5.2 GHz. In the second stage, the backward-wave oscillator (BWO) tube is locked to a harmonic of the Klystron. By changing the BWO and the corresponding elements of the waveguide components, the frequency region from 50 to 120 GHz can be covered. Investigation of the energy spectrum of the synthesizer indicates that the output spectral bandwidth does not exceed 1 kHz. Measurements are carried out with the use of frequency modulation (FM) of the synthesizer radiation. The lines are detected with a room-temperature Schottky diode detector. The long-term frequency stability is completely determined by the stability of the rubidium frequency standard.

For the work here, the spectrometer has been extensively modified as compared to the original version described in Ref. (19). Thus, in place of the selective amplifier in the detection system, a broadband (10–300 kHz) low-noise amplifier was

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used. This permits us to vary the frequency modulation over a broad range and allows the use of frequency modulation not only in the first, but also in the second stage of the PLL, in order to reach the highest resolution with a very small frequency and index of modulation. The Lamb dip in the spectrum of SO_2 was recorded with a width of 0.015 MHz at a frequency around 70 GHz.

After upgrading the hardware and software of the spectrometer, a completely automated procedure was introduced for all the PLL systems. With these upgrades in place, the spectrometer is capable of continuous recording of spectra in all operating frequency regions. Broadband survey scans are carried using comparatively large (up to 50 kHz) frequency steps. In this way, the entire frequency region of the spectrometer (50–150 GHz) can be recorded in several days. These large survey scans are useful in the initial stages of the investigations since they give a general spectral overview. Shortcomings of the survey spectrum consist of some lowering of the precision of the frequency measurements of the transitions (uncertainties from 10 to 50 kHz) and a lower signal/noise ratio. However, since the survey scans are followed by high-resolution measurements, these shortcomings are tolerable.

The complete spectrum of acetic acid in the working region of the spectrometer was recorded in the survey mode with a frequency step of 30 kHz. The uncertainty of these measurements is estimated to be not worse than 10 kHz for an isolated line. This recording of the spectrum was obtained using a single pass absorption cell (20) at room temperature. The pressure in the cell was maintained at a level that insured maximum sensitivity. Under these conditions the width of the lines was near the Doppler limit and did not exceed 0.3 MHz. A total of about 13 000 transitions were recorded, corresponding to various states of CH₃COOH. To increase the frequency measurement accuracy, a number of the transitions were rerecorded at high resolution (Doppler-limited linewidth). Additionally, the frequency range was extended to 155 GHz using a frequency doubler.

III. THEORETICAL MODEL

Rather complete descriptions of the theoretical model used in the present study exist in the literature, since its most recent version has now been applied to a number of molecules containing a C_{3v} internal rotor and an asymmetric C_s molecular frame, e.g., acetaldehyde CH₃CHO (11), methanol CH₃OH (12), methyl mercaptan CH₃SH (21), trifluoropropene CF₃CHCH₂ (22), deuterated acetaldehyde CD₃CHO (23), and several methanol isotopomers (24, 25). We thus give here only the main characteristics.

The Hamiltonian used is the so-called RAM internal-rotation Hamiltonian based on the work of Kirtman (26), Lees and Baker (27), Herbst *et al.* (28), and Liang *et al.* (29). The method takes its name from the choice of axis system (30), the rho axis system, which is related to the principal axis system a, b, c by a rotation chosen to eliminate the $-2FP_{\gamma}\rho_xJ_x$ and

 $-2FP_{\gamma}\rho_{\nu}J_{\nu}$ coupling terms in the kinetic energy operator, where F is the internal rotation constant, P_{γ} is the internal rotation angular momentum, J_x and J_y are the usual x and y components of the global rotation angular momentum, and ρ is a vector that expresses the coupling between the angular momentum of the internal rotation P_{γ} and that of the global rotation J. Rotation to the RAM axis system corresponds to making the new z axis coincident with the ρ vector, since ρ_x = $\rho_v = 0$ by definition. The advantage of the resulting RAM Hamiltonian for computation arises from the fact that all loworder operators containing the torsional angle γ and its conjugate momentum P_{γ} are diagonal in the rotational quantum number K. All operators off-diagonal in K then appear only in the rotational part of the problem. The method starts with a one-dimensional threefold periodic potential function $V(\gamma)$ together with a torsion-rotation kinetic energy operator diagonal in K. In the first step a set of torsional calculations, one for each value of K and Γ (where $\Gamma = A$ or E), is carried out using a 21-function torsional basis set. This basis set is then reduced in size by discarding all but the nine lowest torsional eigenfunctions for given K and Γ . We verified that these two truncations of the Hamiltonian matrix did not modify the energy levels at the level of measurement precision. The retained torsional eigenfunctions are multiplied by symmetrictop rotational functions $|J, K, M\rangle$ to form a basis set which is used to diagonalize, in a second step, zeroth-order asymmetricrotor terms and higher order terms obtained by multiplying torsional operators with rotational operators. (Note, as indicated later in Table 2, that powers of P_{γ} itself, rather than powers of $(P_{\gamma} - \rho J_z)$, are used to construct such higher order operators in this work.)

Acetic acid has three characteristics that the RAM program never had to face before, and this makes it an excellent molecule to test performance of the method in a different limiting case. First, the torsional potential barrier is rather low ($V_3 \approx$ 168 cm⁻¹) so torsional splittings can reach several gigahertz even in the torsional ground state. (From a reduced barrier point of view, however, the torsional problem is somewhere between that for methanol and that for acetaldehyde, since $s \equiv$ $4V_3/9F$ is 6.0 for methanol, 14.5 for acetic acid, and 23.7 for acetaldehyde.) Figure 1 shows a plot of the lowest torsional levels of acetic acid in its threefold potential well, indicating that the $v_t = 2$ level straddles the top of the barrier. (The two lowest small-amplitude vibrations (31), the CO torsion ν_{17} at 534 cm⁻¹ and the CCO deformation ν_{12} at 581 cm⁻¹, are well above the top of the barrier.) To avoid complications from incipient free-rotor states, we have confined our attention in this paper to the $v_t = 0$ and 1 levels, which lie completely below the top of the barrier.

Second, contrary to all previous molecules we have investigated, acetic acid is a fairly heavy, somewhat oblate internal rotor ($\kappa = +0.38$), with the methyl top internal rotation axis perpendicular to the near symmetric-top axis of the molecule, rather than approximately parallel to it. For this reason, K_c will

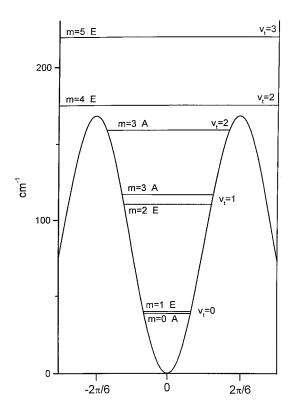


FIG. 1. Low-lying torsional energy levels of acetic acid. The free-rotor quantum number m is given on the left. This quantum number is not well defined for levels below the barrier. The harmonic oscillator vibrational quantum number v_i is given on the right. This quantum number is not well defined for levels above the barrier.

often be a better quasi-quantum number than K_a (though our computer program is set up in a K_a symmetric-top basis set) and transitions will cluster differently than in a near-prolate top molecule. For example, calculations based on the present fit indicate that for J=15, the $J_{J-Kc+1,Kc}$ and $J_{J-Kc,Kc}$ A-species pairs are degenerate to less than 1 kHz for $K_c=15$, 14, and 13 ($K_a=0$, 1, 2, and 3) for both $v_t=0$ and 1. This relatively uncommon very tight clustering behavior prompted the use of the unorthodox $8_{*,8}$ – $7_{*,7}$ and $9_{*,9}$ – $8_{*,8}$ notation in the astronomical searches (I). It also means that our count of 2500 transitions is somewhat misleading, in the sense that one measured frequency can sometimes be counted as four different transitions.

Third, the ratio of the methyl top moment of inertia to that of the rest of the molecule is small. The resultant small value for ρ of 0.08 (compared, for example, to values near 0.3 for acetaldehyde CH₃CHO, 0.8 for methanol CH₃OH, and 0.5 for CD₃CHO), leads to a relatively small coefficient for the coupling term between internal rotation and global rotation. One effect of the small value of ρ is to lengthen the period of the cosine function describing torsional splittings in Eq. [4] of Ref. (30), as well as of the sine function in Eq. [6] and Fig. 2 there, so that for the $F(P_{\gamma} - \rho K)^2$ sign choice in our program, the $+K_a$ E-species levels with $v_t = 0$ ($v_t = 1$) lie below (above)

the $-K_a$ levels in the large interval from $|K_a| = 1$ to $|K_a| = 19$.

It should be noted that for fits of the acetic acid spectrum presented here we have changed from the $F(P_{\gamma} + \rho K)^2$ sign convention used in all previously published fits from the present computer program to the $F(P_{\gamma} - \rho K)^2$ convention. The minus sign arises naturally in the derivation of the Hamiltonian operator when γ is defined to measure the orientation of the top with respect to that of the frame. (See Sections 2IIA and 2IIB of Ref. (32), Section 3 of Ref. (30), and Section II of Ref. (33).) The main consequences of such a sign change in the Hamiltonian operator (if ρ is kept positive) are to change the signs of the $+K_a$, $-K_a$ energy-level labels for E-species transitions in the data set and to change the signs returned by the global least-squares fit for molecular constants multiplying operators which go into their negatives when $\gamma \rightarrow -\gamma$. These consequences can be deduced by noting that the desired sign change can be accomplished formally by changing the sign of the internal rotation angle (33).

The partially resolved hyperfine structure observed for some transitions at the 10-kHz Fourier transform resolution, which presumably arises from spin-spin and/or spin-rotation interactions involving the four hydrogen nuclei, has not been addressed theoretically. As Fig. 2 illustrates, many low J transitions in CH₃COOH involving A-species levels appear as clear weak-strong-weak triplets, whereas the analogous transitions involving E-species levels appear as singlets. As Fig. 2 also illustrates, this same pattern is exhibited by transitions in CH₃CHO, a closely related chemical structure consisting of boson nuclei plus four protons. At present, this empirical regularity has only been used as a consistency check for A/Eassignments. Also, since the frequency of the strongest component is used directly in the fits, the assigned uncertainty of 4 kHz may be somewhat optimistic for these hyperfine-split lines.

IV. ASSIGNMENTS AND FIT

The assignment procedure began by collecting and fitting all previous measurements in the literature for the ground (v_t = 0) and first excited ($v_t = 1$) torsional states (5–10). The predictions obtained were sufficiently good to allow us to assign a number of new transitions, update the fit, and then iterate the procedure through additional cycles. Our confidence in the $v_t = 0$ line assignments is high because (i) the cold (several Kelvin) and very precise NIST Fourier transform measurements can only be assigned to very low J and Ktransitions with $v_t = 0$, (ii) the extensive and precise Kharkov measurements include a large number of four-line loops whose frequencies sum to zero within experimental error, and (iii) the set of assigned ground torsional state transitions, which provide a relatively dense coverage of the rotational quantum number ranges $0 \le J \le 30$ and $0 \le K_a \le 15$, all fit well to a theoretical model which has been thoroughly tested on other 290 ILYUSHIN ET AL.

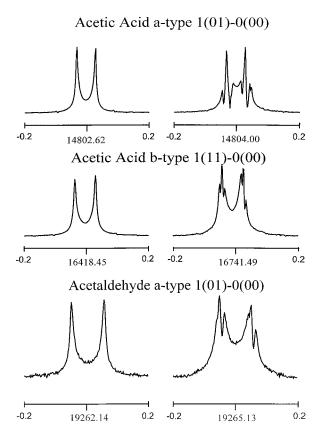


FIG. 2. Six spectral traces illustrating similarities and differences in the hyperfine structure of $J=1 \leftarrow 0$ transitions. Abscissa labels are in megahertz. The ordinate scale is arbitrary intensity units, with the noise level shown in the wings of the lines. The first two rows show the $1_{0,1}$ – $0_{0,0}$ and $1_{1,1}$ – $0_{0,0}$ transitions of acetic acid (with argon carrier). The third row shows the $1_{0,1}$ – $0_{0,0}$ transitions of acetaldehyde (with neon carrier), which clearly resemble their acetic acid counterparts. The left column shows *E*-species transitions, which have no discernible hyperfine structure at the 10-kHz linewidth of the NIST jet-cooled Fourier transform microwave spectrometer. The right column shows *A*-species transitions, all of which appear to have three hyperfine components. The partially resolved hyperfine structure in this figure and the similar hyperfine structure observed for various other *A*-species transitions are not treated in this work. The frequency of the strong central component is used in the fits.

molecules. Our confidence in the $v_t = 1$ assignments and in the full parameter set returned by the fit is not quite so high, because the J and K coverage is still rather poor and there is as yet no direct measurement of the $v_t = 1 \leftarrow 0$ infrared transition frequency, i.e., all parameters must be determined from rotational energy spacings alone.

One of the most difficult problems in attempting to obtain a weighted standard deviation near unity from a global fit of a composite data set is the allocation of appropriate experimental uncertainties. If uncertainties are too lax, the theoretical model appears excellent even when misassignments and/or mismeasurements are present in the fit, leading to gross overconfidence in any calculated transitions. If uncertainties are too tight, the search for useful higher order terms in the model is endless. The weighting strategy chosen for the present work is the

following. Poorly fitting transitions from the older literature, where possibilities of systematic or typographical errors cannot be reexamined, were arbitrarily given zero weight or uncertainties larger than stated in the article. The NIST molecular beam FTMW measurements, which are known from other work to have standard uncertainties of 1 or 2 kHz, have here been given uncertainties of 4 kHz, since we do not expect the Hamiltonian at our present stage of understanding to be accurate to more than a few kilohertz. Most of the Kharkov measurements, which comprise over half of our total data set and which are thought on the basis of a large number of combination difference loops to be accurate to 10 kHz or better, have been given a weight of 20 kHz, again to accommodate model errors. Similarly a number of $v_t = 1$ transitions from Ref. (10), though presumably accurate to 50 kHz, were given a weight of 200 kHz to accommodate model errors. Finally, lines calculated to be unresolved doublets were given weights commensurate with their calculated splittings.

The global fit finally chosen for the present work allowed 34 parameters to vary and gives microwave rms deviations of 41 kHz for 2109 $v_t = 0$ transitions and 93 kHz for 409 $v_t = 1$ transitions with $J \leq 30$, $K_a \leq 15$. The overall quality of the

TABLE 1

Root-Mean-Square Deviations from the Global Fit^a

Number of pa	rameters		34	
Number of lin	ies		2518	
RMS of the 2	109 MW <i>v</i> _t =	= 0 - 0 Lines	0.041	MHz
RMS of the 4	$09 \text{ MW } v_t =$	1 - 1 Lines	0.093	MHz
Sourceb	Range ^c	Linesd	Uncerte	RMSf
	GHz		MHz	MHz
NIST FTMW	8-40	77	0.004	0.004
KHARKOV	49-155	1370	0.020	0.016
VARIOUS		868	0.050	0.044
LILLE	148-250	127	0.100	0.102
KRI71 & VA	N83	72	0.200	0.204
NIST	350-365	4	1.000	0.434

^a Parameter values are given in Table 2. Observed minus calculated residuals are given in Table 3 for $v_t = 0$ lines and in the archived material for $v_t = 1$.

^b Sources for data: KHARKOV, LILLE, NIST = this work; VARIOUS = Refs. (6, 8–10) and this work (Kharkov and NIST non-FT); KRI71 = Ref. (5); VAN83 = Ref. (10).

^c Range containing the measurements in a given row.

^d Number of MW lines in each uncertainty group.

^e One-sigma standard uncertainty in MHz used in the fit, which is sometimes larger than the apparatus capabilities because of model problems or unresolved doublets (see text).

f Root-mean-square deviation in MHz for each group.

TABLI	E 2
Torsion-Rotation Parameter	s Used for the Global Fit

nlma	Operator ^b	Parameter ^b	Value ^c	nlm ^a	Operator ^b	Parameter ^b	Value ^c
220	$\frac{1}{2}(1-\cos 3\gamma)$	V_3	170.217(1)	413	$P_{\gamma}P_a^3$	$\mathbf{k_1}$	-0.1163(7)×10 ⁻⁵
	P_{γ}^{2}	F	5.62160(3)		$P_{\gamma}\{P_a,(P_b^2-P_c^2)\}$	c_4	$0.1744(1) \times 10^{-5}$
211	$P_{\nu}P_{a}$	ρ	0.0719483(3)	404	$-\mathbf{P}^4$	$\mathbf{D_{J}}$	$0.14318(4) \times 10^{-6}$
202	$P_a^{'2}$	A	0.3777291(7)		$-P^{2}P_{a}^{2}$	$ m D_{JK}$	$0.3491(2) \times 10^{-6}$
	P_b^2	\mathbf{B}	0.3166921(2)		$-P_a^4$	D_{K}	$-0.59(6)\times10^{-8}$
	P_c^2	C	0.17766123(7)		$-2P^{2}(P_{b}^{2}-P_{c}^{2})$	$\delta_{ m J}$	$0.5737(2)\times10^{-7}$
	$(P_aP_b+P_bP_a)$	$\mathrm{D_{ab}}$	-0.0040735(3)		$-\{P_a^2,(P_b^2-P_c^2)\}$	δ_{K}	$0.35144(8) \times 10^{-6}$
440	P_{v}^{4}	k_4	$-0.211(1)\times10^{-3}$		$(P_aP_b+P_bP_a)P^2$	D_{abJ}	$-0.328(5)\times10^{-7}$
	$\frac{1}{2}(1-\cos 6\gamma)$	V_6	-6.6390(6)	642	$(1-\cos 6\gamma)P^2$	N_{v}	$0.305(2) \times 10^{-4}$
431	$P_{\nu}^{3}P_{a}$	k_3	$0.287(3) \times 10^{-4}$		$(1-\cos 6\gamma)P_a^2$	K_2	$-0.45(2)\times10^{-4}$
422	$P_{\nu}^{'2}P^{\overline{2}}$	G_{v}	$-0.563(1)\times10^{-5}$		$(1-\cos 6\gamma)(P_b^2-P_c^2)$		$0.133(2) \times 10^{-4}$
	$2P_{y}^{2}(P_{b}^{2}-P_{c}^{2})$	\mathbf{c}_1	$-0.1969(4)\times10^{-5}$	624	$(1-\cos 3\gamma)P^2P_a^2$	k _{5J}	$0.108(3) \times 10^{-7}$
	$(1-\cos 3\gamma)P^2$	$\mathbf{F}_{\mathbf{v}}$	$-0.3550(2)\times10^{-3}$		$(1-\cos 3\gamma)P_a^4$	k _{5K}	$-0.118(8)\times10^{-7}$
	$(1-\cos 3\gamma)P_a^2$	\mathbf{k}_{5}	$0.557(2)\times10^{-3}$		$P_{\gamma}^{2}\{P_{a}^{2},(P_{b}^{2}-P_{c}^{2})\}$		$0.256(4) \times 10^{-9}$
	$(1-\cos 3\gamma)(P_b^2-P_c^2)$		$-0.1912(2)\times10^{-3}$	606	P^6	$\widetilde{\mathrm{H_{J}}}$	$0.285(8) \times 10^{-12}$
	$(1-\cos 3\gamma)(P_aP_b+P_t)$		$-0.22242(3)\times10^{-2}$		$2P^4(P_b^2-P_c^2)$	$h_{ m I}$	$0.125(4)\times10^{-12}$
413	$P_{\nu}P_{a}P^{2}$	L _v	$0.3218(2) \times 10^{-5}$		$P^{2}\{\hat{P}_{a}^{2},(P_{b}^{2}-P_{c}^{2})\}$	h _{JK}	$0.67(2)\times10^{-12}$

^a Notation of Ref. (11); n = 1 + m, where n is the total order of the operator, 1 is the order of the torsional part and m is the order of the rotational part, respectively.

fit is illustrated in Table 1, which gives rms deviations for transitions grouped according to their measurement uncertainties (weight in the fit). Although we fit both A- and E-species transitions simultaneously, we calculate separate rms deviations for A (61 kHz) and E (43 kHz) lines to demonstrate the similar quality of the fit for the two symmetry species. These results represent a very significant improvement over past studies, where the E species could not be reproduced as well as the A species, and overall standard deviations rarely approached experimental precision.

Table 2 presents values and one-sigma standard uncertainties for the 34 parameters used to obtain the fit summarized in Table 1. Before choosing this "best" set of parameters, we examined a large number of other fits. As might be expected (34, 35), parameter values vary significantly (over thousands of standard uncertainties) when fundamental changes in the parameter set are made. For example, V_3 changed from 170.2 to 168.4 cm^{-1} and V_6 changed from -6.6 to -3.2 cm^{-1} when the term $k_7\{(1-\cos 3\gamma), P_{\gamma}^2\}$ was included in the Hamiltonian. Some of these uncertainties in choice of parameter set can be removed by treating more extensive data sets, but some will remain because of arbitrary implicit contact transformation choices (34, 35).

Table 3 presents all $v_t = 0$ transition frequencies included in the fit, together with their assignments, observed minus calculated residuals, and laboratory of origin. When the same transition was measured in more than one laboratory, only the

frequency with the lowest uncertainty was included in the fit in most cases. Agreement between duplicate measurements from the three laboratories contributing new data to this paper was excellent, i.e., mostly within the stated uncertainty limits. Transitions in Table 3 are not ordered by frequency, but are rather grouped into spectroscopic branches, so that model inadequacies can more easily be recognized by examining observedminus-calculated trends along a branch. Ground state transitions of A species are given first. Some branches with $|\Delta K_a|$ 1 are included at appropriate places. Branches with $|\Delta K_c| > 1$ were not systematically searched for in the present work, but branches with $|\Delta K_c|$ = even are forbidden by symmetry for A species. The $v_t = 0$ transitions of E species are given next. A few $|\Delta K_c|$ = even transitions are included, since these are not symmetry forbidden for E species. The smaller number of $v_t =$ 1 transitions (A and E species) are not given here; they can be found in the archived material.

V. DISCUSSION

We present here the first global study of rotational levels with $J \leq 30$ and $K_a \leq 15$ in the lowest two torsional states of acetic acid, using a torsion-rotation Hamiltonian which has proved to be a powerful tool for the analysis of microwave, millimeter-wave, and far-infrared data of internal C_{3v} rotors. This study represents a good starting point for further investi-

^b Notation of Ref. (11). {A, B} = AB + BA. The product of the parameter and operator from a given row yields the term used in the torsion-rotation Hamiltonian, except for F, ρ and A, which occur in the Hamiltonian in the form $F(P_{\gamma} - \rho P_{a})^{2} + AP_{a}^{2}$.

^c Values of the parameters from the fit shown in Table 3. All values are in cm⁻¹, except for ρ which is unitless. Type A statistical uncertainties are shown as one standard uncertainty in the last digit.

Assignments, *Observed Frequencies, *Residuals from the Fit, and Data Sources for Microwave Transitions within the v_t = 0 State TABLE 3

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" Upper and lower state quantum numbers are indicated by ' and ", respectively. Torsion—rotation levels of A species, given in the first part of the table, have a  $\pm$  "parity" label; levels of E species, given in the second part of the table, have a signed  $K_a$  value (30).

^d Data sources: KHARK, LILLE, NIST = this work; KR171 = Ref. (5); WLO88 = Ref. (6); VAN81 = Ref. (8); DEM82 = Ref. (9); VAN83 = Ref. (10).

^b Observed v_i = 0 microwave transitions in MHz, with assigned standard uncertainties in kHz in parentheses, except for (990) which actually means (1000). ° Observed minus calculated residuals (O - C) in MHz.

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gation of torsion-rotation transitions of CH₃COOH for astrophysical use, since it has considerably improved the rms deviations in comparison with previous studies.

The ground torsional state is in very good shape for levels with J up to 30, since a large body of microwave transitions involving these levels can be fit to experimental precision. Our understanding of levels with  $v_t = 1$  must be improved by adding more of the existing pure rotational measurements for this state to the fit, since calculated  $v_t = 1$  transitions at present can sometimes miss their (apparently) corresponding measured lines by several megahertz. (Adding large numbers of such "calculated assignments" to the fit without intermediate fits and other checks is dangerous, so such additions must proceed slowly and carefully.) There is some hope of ultimately including  $v_t = 2$  levels in the fit, since the  $v_t = 2$ , K = 0, A state lies just below, and the K = 0, E state lies just above the top of the barrier in Fig. 1.

It should be noted that a cold molecular beam spectrum of the  $v_t = 1 \leftarrow 0$  fundamental torsional band, which is predicted to lie near 79.1 and 72.8 cm $^{-1}$  for the A and E species, respectively, will eventually be necessary to stabilize and properly constrain the pure torsional parameters F,  $V_3$ ,  $V_6$ , etc. Such parameters are determined here only very indirectly from rotational intervals within each torsional state. Their fitted values are thus very susceptible to large systematic errors arising (i) from slight contamination of rotational-interval calculations by effects from vibrational averaging, (ii) from implicit reduction of the Hamiltonian by contact transformation, and (iii) from other phenomena not considered in the present model. (Surprisingly, however, the value of  $V_3$  determined in this way has been remarkably stable, varying only between 168.0 and 170.2 cm⁻¹ during the last 30 years (5, 6, 8-10).) Attempts to obtain a room-temperature spectrum of the torsional fundamental using a Fourier transform infrared spectrometer have been carried out in several laboratories, but only very weak features can be seen in the predicted regions (36– 38). (It is possible that much of the acetic acid is present in dimeric form at the pressures needed for infrared studies.)

Were it not for the experimental difficulties associated with cooling and resolving the far-infrared torsion–rotation spectrum, acetic acid would be an ideal case for studying pure torsion–rotation interactions above the barrier, since the top of the barrier lies some  $400~{\rm cm}^{-1}$  below the lowest small-amplitude fundamental vibrations  $\nu_{12}$  and  $\nu_{17}$  (31). For this reason, we plan to continue work on its spectrum. An intensity calculation has been carried out, based on the assumption that the components of the permanent electric dipole moment, are driving the torsion–rotation transitions. This calculation will be used (i) to determine which additional weak lines should be searched for, e.g., which A lines with  $|\Delta K_a| \geq 2$ ,  $|\Delta K_c| \geq 3$  have reasonable intensity in this rather asymmetric top ( $\kappa = +0.38$ ) and (ii) to determine if and why branches of the form  $(J+1)_{Ka+1,J-Ka}-J_{Ka,J-Ka+1}$ , for which we have so far found

rather few lines, are predicted to be unexpectedly weak. In addition, we plan to greatly extend our  $v_t = 1$  assignments and to begin including some  $v_t = 2$  lines in the fits.

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