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# Recommended acetylene $12\text{ C } 2\text{ H } 2$ line list in $13.6\mu\text{m}$ spectral region: 2 New measurements and global modeling

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1           **Recommended acetylene  $^{12}\text{C}_2\text{H}_2$  line list in 13.6 $\mu\text{m}$  spectral region:**

2                           **New measurements and global modeling**

3  
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27 **Abstract**

28           Following a previous work on recommended  $^{12}\text{C}_2\text{H}_2$  line list for the 13 – 248  $\text{cm}^{-1}$  and  
29 390 – 634  $\text{cm}^{-1}$  spectral regions, the present work is dedicated on new measurements between  
30 638 and 820  $\text{cm}^{-1}$ . Line intensities have been measured for 18 bands: only four of them were  
31 previously reported. The measurements allowed validating predictions based on the global  
32 modeling of the line positions and intensities performed within the framework of the method of  
33 effective operators. Using the present new measurements as well as previous measurements  
34 from literature, new fittings of the line intensities for the  $\Delta P = 1$  series of transitions have been  
35 performed. A complete calculated line list of 200 bands belonging of the  $\Delta P = 1$  series of  
36 transitions is proposed as supplementary data in order to improve such spectroscopic databases  
37 as HITRAN or GEISA.

38

39

40

## 41 **1. Introduction**

42  
43 The present work is the continuation of a previous paper [1] concerning calculation  
44 between 13 and 248  $\text{cm}^{-1}$  and between 390 and 634  $\text{cm}^{-1}$  with the aim to produce accurate  
45  $^{12}\text{C}_2\text{H}_2$  calculated line positions and intensities. The line list from Ref. [1] has been recently  
46 added in HITRAN2016 database. The calculation is based on the global modeling of the line  
47 positions and intensities within the framework of the method of effective operators [2-4]  
48 applied to various  $\Delta P$  series [1,5-9] ( $P$  being polyad number defined in Section 3). In the  
49 present work new experimental spectra have been recorded and measurements of line  
50 intensities have been performed for 1020 transitions belonging to 18 vibrational bands between  
51 638 and 820  $\text{cm}^{-1}$ . The positions and intensities of transitions (including those of 14 bands  
52 never measured before) are in very good agreement with acetylene line list presented in  
53 calculated acetylene database ASD-1000 [10]. This fact insures that the calculated line list in  
54 the region of the  $\Delta P=1$  series is very close to the measuring data. As consequences, a set of 200  
55 bands has been selected to generate the acetylene calculated line list of 40021 transitions  
56 between 389 and 893  $\text{cm}^{-1}$ . This line list is available to update atmospheric and planetary  
57 databases as HITRAN [11] and GEISA [12]

58  
59 The experimental spectra recorded in MONARIS laboratory and their analysis will be  
60 presented in Section 2. The theoretical approach will be discussed in Section 3. In Section 4,  
61 the line intensity new fit for the  $\Delta P = 1$  series of bands will be described. Section 5 will be  
62 devoted to the description of the recommended  $^{12}\text{C}_2\text{H}_2$  line list and comparisons of calculations  
63 with measurements and databases will be presented in Section 6. Note that the reader is  
64 addressed to color version of this article for a complete readability of most of the figures.

65  
66

## 67 2. New measurements in the 638 – 820 cm<sup>-1</sup> spectral region

68 The previous work [1] was dedicated to  $\Delta P = 0$  series, as well as the beginning of  $\Delta P =$   
69 1 series with very weak bands recorded with absorption paths of around 150 m. To study  
70 weaker bands in the middle of the  $\Delta P = 1$  region (13.6  $\mu\text{m}$ ), such high absorption path is  
71 useless since the spectra will be totally saturated. An absorption path of 25.7 cm has been used  
72 to record three high resolution spectra with pressure of acetylene ranging from  $(2-10)\times 10^{-3}$   
73 atm allowing us to observe 4 bands already studied in literature as well as 14 bands never  
74 studied before. Experimental conditions are summarized in Table 1. For that, the Bruker IFS  
75 125 in MONARIS has been equipped with a KBr beam splitter, and a MCT-D316 detector. A  
76 Globar source has been used. Temperature in the cell is measured with platinum probes inside  
77 the cell (accuracy of  $\pm 0.1\text{K}$ ). Pressure of natural acetylene inside the cell is measured with a  
78 10mbar-full scale Baratron gauge (stated accuracy of  $\pm 0.25\%$ ). A wavenumber calibration has  
79 been performed based on line position of  $^{12}\text{C}_2\text{H}_2$  transitions from HITRAN [11] as etalon. The  
80 average correcting factor  $\varepsilon = (\sigma_{\text{ref}} - \sigma_{\text{obs}})/\sigma_{\text{ref}}$  has been found equal to  $-6.6(1)10^{-7}$  (between  
81 parenthesis is 1 standard deviation 1SD). At  $700\text{ cm}^{-1}$ , it corresponds to a correction of  $-$   
82  $0.46(1)\times 10^{-3}\text{ cm}^{-1}$ . The accuracy of absolute line positions measurements is estimated to be  
83 equal to  $0.1\times 10^{-3}\text{ cm}^{-1}$ .

84 (Table 1)

85  
86 Figure 1 shows a portion around  $705-706\text{ cm}^{-1}$ , where the strongest saturated line is the  
87  $P_{ee}(10)$  transition of the  $\nu_5^1$  cold band of  $^{12}\text{C}_2\text{H}_2$ . Around  $705.243\text{ cm}^{-1}$  the same transition is  
88 observed but corresponds to  $^{12}\text{C}^{13}\text{CH}_2$  isotopologue (marked with red stick in the upper panel  
89 of Fig. 1). Although only  $^{12}\text{C}_2\text{H}_2$  transitions have been studied in the present work the  
90 parameters of such  $^{12}\text{C}^{13}\text{CH}_2$  transitions have been fitted together with nearby lines. HITRAN  
91 [11] and GEISA [12] databases contain the 5 most intense vibrational bands in the studying  
92 region coming from the Ref. [13,14]. All these bands were reconsidered here except the  
93 strongest  $\nu_5^1$  band which is saturated in our spectra. Note also that the two bands  $(\nu_4 + \nu_5)_+^0 - \nu_4^1$   
94 and  $(\nu_4 + \nu_5)_-^0 - \nu_4^1$  present in HITRAN and GEISA from Ref. [13] correspond to only one band  
95 noted “00011 1-1 \_ 00010 1 0” in the present work using vibrational assignment based on  $V_1 V_2$   
96  $V_3 V_4 V_5 \ell_4 \ell_5$  quantum numbers (see Section 3.2 of Ref. [1] for more details).

97 The peak to peak signal-to-noise ratio of the spectra is almost constant with the  
98 wavenumber and is around 70. As it can be guessed in Fig. 1, a multiplicative channel  
99 spectrum is observed on all experimental spectra. This channel spectrum was modelled locally

100 as a background adjusted by a second order polynomial function within spectral domains  
101 around 0.04-0.1  $\text{cm}^{-1}$ .

102 (Figure 1)

103  
104 The multispectrum fitting procedure of Ref. [15] was used fixing the self-broadening  
105 coefficients at the values of Ref. [16] and using the Voigt profile. The apparatus function has  
106 been calculated with the maximum of optical path difference equal to 225 cm and the nominal  
107 value of the radius of the beam inside the interferometer equal to 1.0 mm together with the  
108 focal distance equal to 418 mm. An example of the result of the multispectrum fit is given in  
109 Fig. 2 for the  $P_{ee}$  (24) transition of the  $2\nu_5^2$  band of  $^{12}\text{C}_2\text{H}_2$  between 675.809 and 675.849  $\text{cm}^{-1}$ .  
110 Taking into account the signal to-noise ratio (which is close to 70), the residuals of the fit do  
111 not show any strong characteristic signature despite a kind of asymmetry in the profile that  
112 gives residuals slightly larger than the 1.5 % peak to peak noise of the experimental spectra.

113

114 (Figure 2)

115

116 Absolute line intensities have been retrieved for 1220 transitions involving 18 vibrational  
117 bands. The accuracy of line intensities has been estimated to be around 5% for transitions deep  
118 enough in recorded experimental spectra (with intensities around  $10^{-20}$ – $10^{-21}$   
119  $\text{cm}^{-1}/(\text{molecule}\cdot\text{cm}^{-2})$ ) whereas the accuracy may reach up to 10% for weaker transitions (with  
120 intensities around  $10^{-22}$ – $10^{-23}$   $\text{cm}^{-1}/(\text{molecule}\cdot\text{cm}^{-2})$ ). A sample of the results for the  $2\nu_5^0 - \nu_5^1$   
121 band is given in Table 2 (the whole set of the measurements is given in Supplementary material  
122 1). A summary of the measurements performed in this work is presented in Table 3 for the 18  
123 studied bands.

124

125 (Table 2)

126 (Table 3)

127

128 **3. Theoretical approach**

129 Measured line intensities were included in the simultaneous fit of the effective dipole  
 130 moment parameters using approach describing in many previous publications (see for example  
 131 [1-5]). The most important points of the approach are given below. The probability of a  
 132 transition between two states  $W_{b\leftarrow a}$  can be deduced from every line intensity expressed in  $\text{cm}^{-1}$   
 133  $^1/(\text{molecule}\cdot\text{cm}^{-2})$  using following conventional formula:

134

$$135 \quad S_{b\leftarrow a}(T) = \frac{8\pi^3 g_i \sigma_{b\leftarrow a}}{3hcZ_{\text{tot}}(T)} e^{-\frac{hcE_a}{k_B T}} \left(1 - e^{-\frac{hc \sigma_{b\leftarrow a}}{k_B T}}\right) W_{b\leftarrow a}, \quad (1)$$

136

137 where  $h$  is the Planck's constant equal to  $6.6260755 \times 10^{-27} \text{erg}\cdot\text{s}$  ( $1 \text{ erg} = 10^{-7} \text{ J}$ );  $c$  is the vacuum  
 138 velocity of light equal to  $2.99792458 \cdot 10^{10} \text{ cm s}^{-1}$ ;  $\sigma_{b\leftarrow a}$  is the transition wavenumber in  $\text{cm}^{-1}$ ;  
 139  $E_a$  is the energy of the lower state (ground state) in  $\text{cm}^{-1}$ ,  $k_B$  is the Boltzmann's constant equal  
 140 to  $1.380658 \times 10^{-16} \text{erg}\cdot\text{K}^{-1}$ ;  $g_i$  is the statistical weight due to nuclear spin of the lower levels  
 141 (equal 3/1 for symmetry  $a/s$ ), and  $T$  is the temperature of the measurements which was 296 K  
 142 in our case. The total partition function  $Z_{\text{tot}}$  used in this work is equal to 412.45 at 296 K [17].  
 143 The probability of a transition  $W_{b\leftarrow a}$  is related to the dipole moment matrix element by the next  
 144 expression ( $m$  is the projection of the angular moment on the  $Z$  axis of the space fixed system  
 145 of axes):

146

$$147 \quad W_{b\leftarrow a} = 3 \sum_{mm'} |\langle \Psi_b | M_Z | \Psi_a \rangle|^2. \quad (2)$$

148

149 In Ref. [4] the eigenfunctions of the effective Hamiltonian of acetylene molecule have been  
 150 obtained in the form of expansion over products of harmonic oscillator and rigid rotator  
 151 wavefunctions:

152

$$153 \quad \Psi_{NJMm\epsilon}^{\text{eff}} = \sum_{V_1 V_2 V_3 V_4 V_5 \ell_4 \ell_5} C_{NJ\epsilon}^{V_1 V_2 V_3 V_4 V_5 \ell_4 \ell_5} |V_1 V_2 V_3 V_4 V_5 \ell_4 \ell_5 J m K \epsilon\rangle, \quad (3)$$

154 where  $N$  identify the eigenstate inside of the polyad of interacting vibrational levels,  $V_i$  are the  
 155 harmonic oscillator quantum numbers,  $\ell_i$  are the vibrational angular moments of the degenerate  
 156 oscillators,  $J$  – angular moment quantum number,  $K$  is the projection of the angular moment on

157 the axis of the molecule,  $\varepsilon$  is the Wang combination symmetry ( $e/f$ ) and  $C$  are the mixing  
 158 coefficients. The sum is performed over all interacting basic functions forming one polyad  
 159 which is defined by polyad number  $P$ . In the case of acetylene molecule, the harmonic  
 160 oscillator quantum numbers of the interacting basic functions should satisfy the equation:

$$161 \quad P = 5V_1 + 3V_2 + 5V_3 + V_4 + V_5 = \text{const}, \quad (4)$$

162 which expresses the approximate relations between normal mode frequencies in this molecule.

163 In order to use these functions in our calculation the dipole moment operator was transformed  
 164 to the effective dipole moment operator using the same unitary transformation as the one  
 165 transforming vibrational-rotational Hamiltonian to the effective:

$$166 \quad W_{N'J'_{\varepsilon'} \leftarrow NJ_{\varepsilon}} = 3 \sum_{mm'} \left| \langle \Psi_{N'J'm_{\varepsilon'}}^{eff} | M_Z^{eff} | \Psi_{NJm_{\varepsilon}}^{eff} \rangle \right|^2 =$$

$$167 \quad = 3 \sum_{mm'} \left| \sum_{\mathbf{V}, \mathbf{V}', \boldsymbol{\ell}, \boldsymbol{\ell}'} C_{N'J'_{\varepsilon'}}^{\mathbf{V}, \boldsymbol{\ell}'} C_{NJ_{\varepsilon}}^{\mathbf{V}', \boldsymbol{\ell}} \langle V', \boldsymbol{\ell}' J' K' m'_{\varepsilon'} | M_Z^{eff} | V \boldsymbol{\ell} J K m_{\varepsilon} \rangle \right|^2, \quad (6)$$

168 where we denoted for simplicity the set of the harmonic oscillator quantum numbers  $V_i$  by  
 169 vector  $\mathbf{V}$  and the vibrational angular momentum quantum numbers  $\ell_i$  by vector  $\boldsymbol{\ell}$ . The sum over  
 170 these vectors assumes the sum over every quantum numbers separately, but satisfying Eq. (4).  
 171 After developing effective dipole moment operator in series the calculations lead to the next  
 172 expression for the probability of the transition:

$$173 \quad W_{N'J'_{\varepsilon'} \leftarrow NJ_{\varepsilon}} = (2J+1) \left| \sum_{\mathbf{V}, \mathbf{V}', \boldsymbol{\ell}, \boldsymbol{\ell}'} C_{N'J'_{\varepsilon'}}^{\mathbf{V}, \boldsymbol{\ell}'} C_{NJ_{\varepsilon}}^{\mathbf{V}', \boldsymbol{\ell}} \sqrt{f_{\Delta \mathbf{V}}^{\boldsymbol{\ell}}(\mathbf{V}, \boldsymbol{\ell})} (1 + |\delta_{\ell_4, 0} \delta_{\ell_5, 0} - \delta_{\ell_4', 0} \delta_{\ell_5', 0}|) \times \right.$$

$$174 \quad \left. \times \Phi_{\Delta J \Delta K}(J, K) M_{\Delta \mathbf{V}}^{\boldsymbol{\ell}} \left( 1 + \sum_i K \Delta V_i^{\boldsymbol{\ell}} V_i + \sum_{i=4,5} \alpha^{\Delta V_i^{\boldsymbol{\ell}}} (2\ell_i \Delta \ell_i + 1) + F_{\Delta J \Delta K}^{\Delta \mathbf{V}, \boldsymbol{\ell}}(J, K) \right) \right|^2. \quad (7)$$

175 The square of the function  $\Phi_{\Delta J \Delta K}(J, K)$  multiplied by factor  $(2J+1)$  gives the Hönl-London factor  
 176 in the case of isolated transitions. For  $\Delta K=0, \pm 1$  this function coincide with Clebsch-Gordan  
 177 coefficients and for  $\Delta K=\pm 2, \pm 3$  can be found in Ref. [5,18]. The function  $f_{\Delta \mathbf{V}}^{\boldsymbol{\ell}}(\mathbf{V}, \boldsymbol{\ell})$  is the  
 178 square of vibrational matrix element and can be found in Ref. [5]. The combination of the  
 179 Kronecker symbols appears due Wang basis functions and provides 2 when one of the basic  
 180 functions has both vibrational angular moments equal zero. The Herman-Wallis type factor  
 181  $F_{\Delta J \Delta K}^{\Delta \mathbf{V}, \boldsymbol{\ell}}(J, K)$  describes the rotational dependence of the effective dipole moment operator. For  
 182  $\Delta K=\pm 1$  it can be written as follows:



183 for  $Q$  branch:

$$184 \quad F_{\Delta J \Delta K}^{\Delta V \Delta \ell}(J, K) = -\frac{1}{2} b^{\Delta V f \Delta \ell} (2K \Delta K + 1) + d^{\Delta V f \Delta \ell} \left[ J(J+1) - K^2 - \Delta K \left( K + \frac{\Delta K}{2} \right) \right], \quad (8)$$

185 for  $P$  and  $R$  branches:

$$186 \quad F_{\Delta J \Delta K}^{\Delta V \Delta \ell}(J, K) = -\frac{1}{4} (d^{\Delta V f \Delta \ell} - d^{\Delta V f \Delta \ell}) - \frac{1}{2} (b^{\Delta V f \Delta \ell} + d^{\Delta V f \Delta \ell}) (2K \Delta K + 1) - d^{\Delta V f \Delta \ell} K^2 +$$

$$187 \quad + b^{\Delta V f \Delta \ell} m + d^{\Delta V f \Delta \ell} m^2 + \frac{1}{2} (d^{\Delta V f \Delta \ell} - d^{\Delta V f \Delta \ell}) m (2K \Delta K + 1). \quad (9)$$

188 In the case  $\Delta K=0$  for  $P$  and  $R$  branches, the Herman-Wallis type factor  $F_{\Delta J \Delta K}^{\Delta V \Delta \ell}(J, K)$  is:

$$189 \quad F_{\Delta J \Delta K=0}^{\Delta V \Delta \ell}(J, K) = b^{\Delta V f \Delta \ell} m + d^{\Delta V f \Delta \ell} [J(J+1) - K^2 + m]. \quad (10)$$

190  
 191 The  $m$  quantity in the Eqs. (9-10) and corresponds to  $m = -J, J+1$  for  $P, R$  branches  
 192 respectively, and should not be confounded with the angular momentum projection quantum  
 193 number used in Eqs. (2,3,6). The expression  $F_{\Delta J \Delta K}^{\Delta V \Delta \ell}(J, K)$  for the  $Q$  branch ( $\Delta K=0$ ) depends from  
 194  $\ell_4$  and  $\ell_5$  values and can be found in Ref. [3]. The set of the effective dipole moment  
 195 parameters:  $M_{\Delta V}^{\Delta \ell}, K_{\Delta V}^{\Delta \ell}, a_{\Delta V}^{\Delta \ell}, b_{\Delta V}^{\Delta \ell}, d_{\Delta V}^{\Delta \ell}, d_{\Delta J}^{\Delta \ell}$  determines the intensity distribution in  
 196 the spectral domain defined by the value of  $\Delta P$ . Note, that the effective dipole moment  
 197 parameters are the same for the vectors  $\Delta V$  and  $\Delta \ell$  differing only by signs except parameter  
 198  $a_{\Delta V}^{\Delta \ell}$  which changes the sign when the sign of the vector  $\Delta \ell$  changes.

199

200

#### 201 4. Line intensity fit ( $\Delta P = 1$ series of transitions)

202

203 Based on the new measurements performed in this work (Section 2) and on the  
204 measurements published earlier [1,13,14], 14 effective dipole moment parameters have been  
205 fitted for the  $\Delta P=1$  series of transitions to 1923 measured line intensities belonging to 28  
206 different bands-(see Table 5). Most of the bands measured in this work (13 from 18) correspond  
207 to  $\Delta V_5 = 1$ , with  $\Delta \ell_4 = 0$  and  $\Delta \ell_5 = 1$ , but also with  $\Delta \ell_4 = -2$  and  $\Delta \ell_5 = +1$  (3 bands), or with  
208  $\Delta \ell_4 = -2$  and  $\Delta \ell_5 = +3$  (1 band), or with  $\Delta \ell_4 = +2$  and  $\Delta \ell_5 = +1$  (1 band). No any new effective  
209 dipole moment parameter was required to reproduce the new measurements. The set of the  
210 fitted effective dipole moment parameters for  $\Delta P = 1$  is presented in Table 4 and the statistics  
211 of the fit is given in Table 5. For some transitions (weakest or blended), measurements of this  
212 work or from Ref. [1,13,14] become less accurate and can be off by more than 10 % from  
213 calculations. Most of these measurements have been excluded from the fit of the parameters of  
214 Table 4 (representing 424 measurements from a total number of measurements equal to 2347).  
215 The present global model (using newly fitted parameters, see Table 4) reproduced 76% (1455  
216 measurements) of the measurements included in the effective dipole moment parameters fit  
217 within  $\pm 5\%$ , and 22% of measurements (430 measurements) within  $\pm 5-10\%$ . The  
218 eigenfunctions of our global effective Hamiltonian [4] were used in the fit. The weighted  
219 dimensionless standard deviation  $\chi = 0.75$  and the root mean squares of the residuals  
220 RMS=4.4% were obtained in result of fit. The effective dipole moment parameters retrieved in  
221 this work are consistent with previous ones except for the  $b_J$  parameter for  $\Delta V_4 = 0$ ,  $\Delta V_5 = 1$ ,  
222  $\Delta \ell_4 = 0$ , and  $\Delta \ell_5 = 1$  that converges to  $-0.49(3) \times 10^{-3}$  in the present fit instead of  $-0.76(5) \times 10^{-3}$   
223 and  $-0.72(5) \times 10^{-3}$  in Ref. [1] and Ref. [4] respectively (see Table 4). For bands measured in  
224 the present work, the 3 sets of effective dipole moment parameters in Table 4 lead to similar  
225 calculations able to reproduce the intensities of transitions never measured before.

226

(Table 4)

227

(Table 5)

228 Examples of calculations and measurements for two newly measured bands are given in  
229 Figs. 3-4. Similar graphic comparisons are available for all measured bands in Supplementary  
230 material 2. As observed in Figs. 3-4, the differences between different calculations are  
231 negligible. Both calculations from effective parameters of Refs. [1,4] and of this work (see  
232 Table 4) are similar and reproduce the rotational dependence of  $R^2$  for the 14 bands newly  
233 observed in this work. As observed in Figs. 3-4, the rotational dependence of measured  $R^2$   
234 values is quite smooth and well reproduced by all calculations (all calculations may not always

235 be distinguished because of the scale and of the similar calculated values). For some branches,  
236 one can clearly see that the measured rotational dependence of  $R^2(m)$  cannot be reproduced by  
237 Herman-Wallis factors.

238 (Figure 3)

239 (Figure 4)

240

## 241 **5. Recommended acetylene ( $^{12}\text{C}_2\text{H}_2$ ) line list for databases**

242

243 Using the effective Hamiltonian parameters of Ref. [4] and effective dipole moment  
244 parameters (see Table 4) of  $\Delta P = 1$  series, the acetylene ( $^{12}\text{C}_2\text{H}_2$ ) line list has been generated in  
245 the studding region with intensity cut off  $10^{-28} \text{ cm}^{-1}/(\text{molecule.cm}^{-2})$ . The spectral lines in this  
246 model do not have a unique vibrational labeling for all transitions belonging to a same band,  
247 but two principal contributors are given for upper and lower levels of transitions which can  
248 change inside a band. In order to have consistent vibrational labelling for a same series of  
249 transitions (with  $P$ ,  $Q$  and  $R$  branches), an algorithm has been developed to reveal levels  
250 crossings. Indeed, as pointed out in Ref. [1], for some bands the main contributor to the  
251 eigenfunctions can change within a same series of transitions (band). When ro-vibrational  
252 levels of different series of transitions, considering as  $J$  function, approach, some levels  
253 crossing can appear. As performed in Ref. [1], the vibrational assignment of the lowest  $J$   
254 transitions has been used to name the band. Inside a series of transitions, when crossings  
255 appear, the algorithm is looking for the next transition belonging to the series using such  
256 criteria as the difference of wavenumbers between transitions and the two largest contributions  
257 of the basic functions (for the upper and lower levels). Such transitions are noted with a star in  
258 the Table 6 and Supplementary material 3. The fact that the main contributor to the  
259 eigenfunctions is changing progressively (slowly or quickly depending on bands) affect line  
260 intensities in an unusual way so that Herman-Wallis factors are unable to reproduce the  
261 rotational dependence of transition dipole moment squared  $R^2$ . This problematic has been  
262 pointed out for example in Ref. [19] for  $\nu_1 + \nu_3 + 2\nu_4^0$  band (see Fig. 3 of Ref. [19]) and in Ref.  
263 [1] for 2 bands (see Figs. 10-11 of Ref. [1]) where the Herman-Wallis factors were not adapted  
264 to fit such unusual rotational dependence. Interesting case of band crossing is also presented in  
265 Ref. [20]. In the present work, several newly observed bands demonstrate such smooth  
266 rotational dependence of  $R^2$  and are well predicted by previous calculations [1,4]. The effects of  
267 crossing levels can be drastic for line intensities calculation. The case of the 00031 1 1 – 00030  
268 1 0 band is discussed as an example. The line intensities and transition dipole moment squared

269 values in the  $Q_{fe}$  branch are plotted in Figs. 5 and 6 respectively. Extract of the whole  
270 calculation (available as Supplementary material 4) with the two main contributors to the  
271 vibrational levels is given in Table 6 for the  $Q_{fe}$  branch of the 00031 1 1 – 00030 1 0 band. The  
272 first crossing appears in the  $Q_{fe}$  branch at  $J$  equal 17, the first contributor in the upper state  
273 becomes 00031 1-1 instead of 00031 1 1. Then at  $J$  equal 18, the first contributor in the upper  
274 state changes again to become 00031 3 1 for the rest of the branch. Such crossings led to a  
275 change of slope for the  $R^2$  values inside the branch (see Fig. 6). As written before, such  
276 rotational dependence within a branch cannot be correctly fitted by Herman-Wallis factors. In  
277 Fig. 5 one can observe the line intensities getting stronger after the crossing point ( $J=18$ , see  
278 Fig. 5). One can also notice in Fig. 5 the alternation of intensities versus  $J$  with a ratio 3:1 for  
279  $^{12}\text{C}_2\text{H}_2$  (due to the nuclear spins for symmetric isotopologues).

280 (Figure 5)

281 (Figure 6)

282 (Table 6)

283 From the complete calculation, 200 series of transitions forming vibrational bands have  
284 been defined and selected for databases. The whole line list contains 41021 transitions. Two  
285 cut-off procedures have been applied to select transitions. First a line intensity cut-off equal to  
286  $10^{-28} \text{ cm}^{-1}/(\text{molecule.cm}^{-2})$  at 296K has been used. Then, only bands with a sum of line  
287 intensities greater than  $10^{-24} \text{ cm}^{-1}/(\text{molecule.cm}^{-2})$  at 296K have been selected. Note that the  
288 sum of line intensities has been halved when the band is affected by  $\ell$ -type doubling (leading  
289 to two series of  $P$ ,  $Q$  and  $R$  branches). A summary of the present calculation for the 30  
290 strongest bands is given in Table 7. The whole summary for the 200 bands calculated in the  
291 recommended line list is given as Supplementary material 4. Note, the recommended line list  
292 includes also most of bands proposed in Ref. [1] and already in HITRAN2016. These bands are  
293 marked by “H” letter in the Table 7 and corresponding Supplementary material 4. Because of  
294 the line intensity cut-off procedures performed in the present work, the 7 weakest bands from  
295 Ref. [1] are not included in the present line list.

296

297 (Table 7)

298 For all 200 bands a line list in HITRAN format has been generated and is available in  
299 Supplementary material 5 to this paper. As discussed in Section 6, the line positions from Refs.  
300 [21,22] present in HITRAN2016 have been used (when available) for the 5 strongest bands  
301 using same error code than in HITRAN2016: 4 (between  $0.1 \times 10^{-3}$  and  $1 \times 10^{-3} \text{ cm}^{-1}$ ). For other

302 transitions, the calculated line positions of the present work (coming from effective  
 303 Hamiltonian parameters of Ref. [4]) have been used with an error code equal to 3  
 304 (corresponding to  $(1 - 10) \times 10^{-3} \text{ cm}^{-1}$ ) for transitions with  $J$  lower or equal to 40 and with an  
 305 error code equal to 2 (corresponding to  $(10 - 100) \times 10^{-3} \text{ cm}^{-1}$ ) for transitions with  $J$  higher than  
 306 40 (see discussion in Section 6). The set of effective dipole moment parameters from the  
 307 present work (see Table 4) has been used to generate line intensities of all bands except the  
 308 cold  $\nu_5^1$  band for which the set of effective dipole moment parameters from Ref. [1] has been  
 309 used in order to be more consistent with measurements (see discussion in Section 6). For line  
 310 intensities a code 5 (between 5 and 10%) was used for the 18 measured bands (noted “Obs” in  
 311 Table 7), whereas for a predicted band the error code 4 ( $\geq 10\%$  and  $< 20\%$ ) was chosen allowing  
 312 to distinguish easily predicted and measured bands. Broadening coefficients (self and air),  
 313 temperature dependence of air-broadening coefficients, air-shifting coefficients and associated  
 314 error codes have been added following recommendations of Ref. [23] included in  
 315 HITRAN2004 edition [24]. However, the error codes of air- and self-broadening coefficients  
 316 have been degraded from 6 (between 2 and 5%) to 4 (between 10-20%) when  $J \geq 35$  since a  
 317 constant estimated value is used. Let us mention that in HITRAN 2016, error codes for  
 318 broadening coefficients are all fixed to 6 even for transitions with  $J$  greater than 35.

319 Note that the vibrational labeling used in the present work has been described in Section  
 320 3.2 of Ref. [1]. We urge spectroscopic databases to note the fact that using  $V_1 V_2 V_3 V_4 V_5 \ell_{\text{tot}}$   
 321 quantum numbers instead of  $V_1 V_2 V_3 V_4 V_5 \ell_4 \ell_5$  quantum numbers will lead to various  
 322 transitions with identical assignments. The FORTRAN format A15 for vibrational assignment  
 323 should change to (1X,3I1,4I2,A1,I1,A1) respectively for  $V_1, V_2, V_3, V_4, V_5, \ell_4, \ell_5$ , quantum  
 324 numbers, the  $u/g$  symmetry, a ranking number (noted  $r$  in HITRAN, see Table 3 of Ref. [24]),  
 325 and the  $+/-$  symmetry.  $V_4$  and  $V_5$  being the lowest energy vibrational modes, two digits should  
 326 be reserved for these modes for the possibility to have  $V$  values greater than 9. The ranking  
 327 number is not used in the present line list and should not be useful anymore.

328 A sample of synthetic spectra using either HITRAN2016 or the present recommended  
 329 line list are compared to experimental spectrum 3 in Fig. 7 between  $703$  and  $706 \text{ cm}^{-1}$ . One can  
 330 notice the many transitions of  $^{12}\text{C}_2\text{H}_2$  missing in HITRAN2016 [11] and GEISA2015 [12] but  
 331 present in our line list. One can also observe some missing absorption features due to  $^{12}\text{C}^{13}\text{CH}_2$   
 332 isotopologue noted by symbols in Fig. 7 and assigned in Ref. [25].

333 (Figure 7)

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## 335 6. Comparisons with spectroscopic databases

336 Since our aim is to update line positions and intensities already present in atmospheric  
337 databases and upgrade the line list with many hot bands, comparisons have been performed  
338 with HITRAN2016 edition. HITRAN2016 [11] or GEISA2015 [12] have same data for  $^{12}\text{C}_2\text{H}_2$   
339 line positions and intensities of strong bands from Refs. [13,14], except for weak bands from  
340 Ref. [1] included in HITRAN2016 edition [11]. Calculations performed in Ref. [1] for the 29  
341 bands in  $\Delta P = 1$  spectral region, and in the present work are based on same formalism [4] so no  
342 comparison is performed. Note that none of the bands measured in the present work is in  
343 common with the bands studied in Ref. [1]. Comparisons with EXOMOL database for  $^{12}\text{C}_2\text{H}_2$   
344 [26] are also performed in term of line positions and intensities.

345 In the  $\Delta P = 1$  absorption region, line intensities of the 5 strongest bands at room  
346 temperature present in HITRAN are coming from Refs. [13,14]. Herman-Wallis factors and  
347 vibrational transition dipole moments squared from Refs. [13,14] have been used to generate  
348 calculated line intensities for updating HITRAN2004 edition [24]. Extrapolations for high  $J$ -  
349 values in HITRAN were performed using fixed  $R^2$  values equal to the calculated one for the last  
350 measured transition in the branch. The present global calculation is consistent with HITRAN  
351 line intensities and improves extrapolations performed in HITRAN with fixed transition dipole  
352 moment squared values. Examples for the  $\nu_5^1$  and  $2\nu_5^0 - \nu_5^1$  bands are given in Figs. 8-9  
353 respectively. Figure 9 shows the very good consistency of present measurements and those  
354 from Ref. [13]. No measurement has been performed in this work for the strong  $\nu_5^1$  band, but  
355 calculations and measurements from literature plotted in Fig. 8 shows a slight discrepancy (up  
356 to 4% for highest  $J$  values) between measurements and calculations in the  $R$ -branch, especially  
357 with parameters of the present work (see Table 4). The  $R^2$  values deduced from EXOMOL line  
358 intensities are also given in Figs. 8 and 9. One can notice that EXOMOL line intensities worse  
359 reproduce measurements showing inaccurate rotational dependences especially for  $P$  and  $R$   
360 branches. The whole set of graphic comparisons is available in Supplementary material 2.

361 (Figure 8)

362 (Figure 9)

363 Line positions of the 5 strongest bands from HITRAN2016 coming from calculation of  
364 Refs. [21,22] (believed to be a reference in the  $\Delta P = 1$  region for line positions) were compared  
365 with ASD-1000 calculated line positions [10] and with present measurements. Comparisons  
366 plotted in Fig. 10 versus line intensity show that for the weakest transitions the differences  
367 between HITRAN and ASD-1000 calculation can reach  $\pm 0.03 \text{ cm}^{-1}$  while present  
368 measurements and HITRAN2016 positions are in very good agreement. In order to better

369 understand the situation, the differences have been plotted versus  $m$  for the 00001 0 1 – 00000  
370 0 0 and 00011 1 1 – 00010 1 0 bands in Fig. 11 and 12 respectively, including also comparisons  
371 with EXOMOL line positions [26]. One can clearly observe in these figures increasing of  
372 deviations of ASD-1000 and EXOMOL line positions from HITRAN values for large rotational  
373 quantum numbers  $J$ . Moreover, while for the 00001 0 1 – 00000 0 0 band (Fig.11) the  
374 differences increase on average in the same way for ASD-1000 and EXOMOL, for the 00011 1  
375 1 – 00010 1 0 band the trend is opposite (Fig.12). In term of line positions, the global model [4]  
376 does not allow to reach the accuracy of more simple models as those from Refs. [21,22] (used  
377 in HITRAN for the 5 strongest bands). These models consider several resonance interactions  
378 between ro-vibrational states and can well describe the rotational dependence of the energy of  
379 several interacting states. The models used in Refs. [21,22] allowed to reach estimated accuracy  
380 equal to  $\pm 0.0001 \text{ cm}^{-1}$  [21]. As consequences the line positions in HITRAN have been kept in  
381 our recommended line list for the 5 strongest bands as discussed in Section 5.

382

383

(Figure 10)

384

385

(Figure 11)

386

387

(Figure 12)

388 **6. Conclusion**

389           Recent measurements from Ref. [1] and from the present work allowed to demonstrate  
390 the predictive capabilities of global calculation [4] for  $\Delta P=1$  series of bands. This calculation  
391 has been used to generate a complete line list in HITRAN format for  $^{12}\text{C}_2\text{H}_2$  isotopologue  
392 between 389 and 893  $\text{cm}^{-1}$ . The cut-off procedures used to select the most intense bands will  
393 allow to model acetylene absorption features with better accuracy for atmospheric temperature  
394 spectra, but also to model higher temperature spectra since the recommended line list contains  
395 many hot bands. The present recommended line list can be considered as a revised selection of  
396 bands from the high-temperature acetylene spectroscopic databank (ASD-1000) [10] in the  $\Delta P$   
397 = 1 spectral region, adapted for spectroscopic databases such as HITRAN or GEISA. By  
398 performing measurements in higher  $\Delta P$  spectral regions for weaker bands never measured, the  
399 validation of the predictive capabilities of global calculation for higher  $\Delta P$  absorption regions is  
400 planned to improve higher spectral regions in spectroscopic databases.

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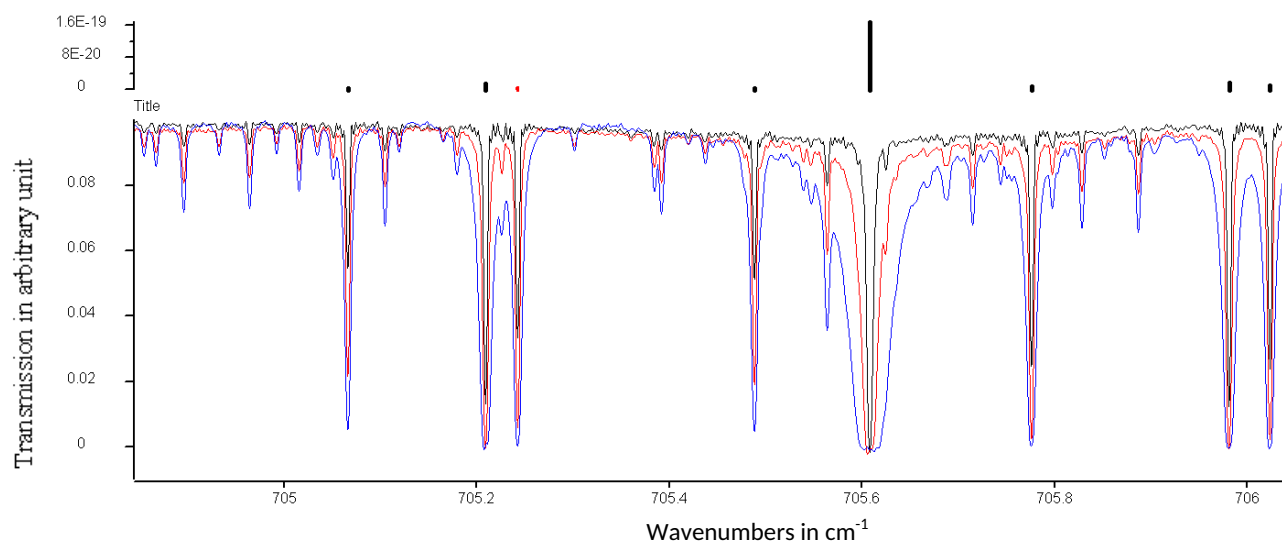


Fig. 1. Spectral range around 705-706  $\text{cm}^{-1}$ . Upper panel is a stick plot of HITRAN 2016 [11] for  $^{12}\text{C}_2\text{H}_2$  (black sticks) and  $^{12}\text{C}^{13}\text{CH}_2$  (red sticks). The line intensities (in log scale) are in  $\text{cm}^{-1}/(\text{molecule}\cdot\text{cm}^{-2})$ . In the lower panel the 3 experimental spectra are plotted (see experimental conditions in Table 1).

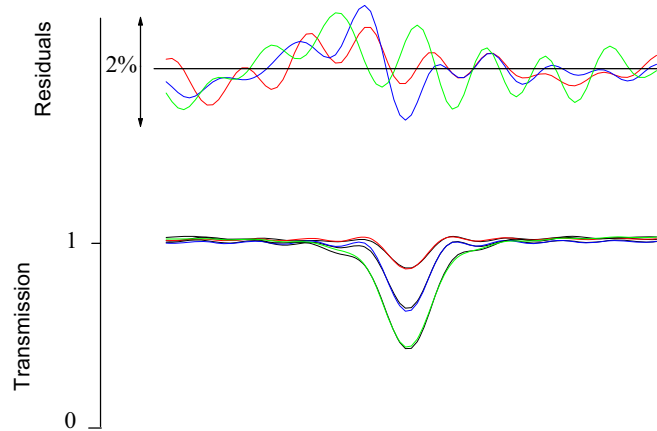


Fig. 2. Simultaneous fit of the  $P_{ee}$  (24) transition of the 00002 0 2 – 00001 0 1 band ( $2\nu_5^2 - \nu_5^1$ ) between 675.809 and 675.849  $\text{cm}^{-1}$ . In the lower panel are plotted the measured spectra #1-3 (in black) and the respective simulated spectra (red for spectrum #1, blue for # 2, green for #3) using multispectrum fitting procedure. In the upper panel are given the residuals of the fit where the same set of colors been used.

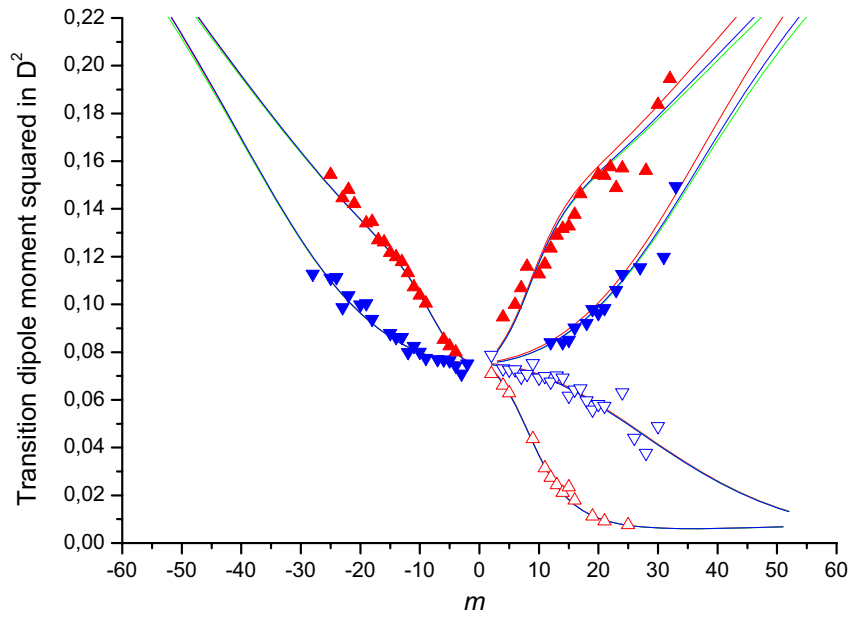


Fig. 3. Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the 00021 2-1 - 00020 2 0 band plotted versus  $m$  ( $m = -J, J, J+1$  for  $P, Q, R$  branch respectively). Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas blue triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4).

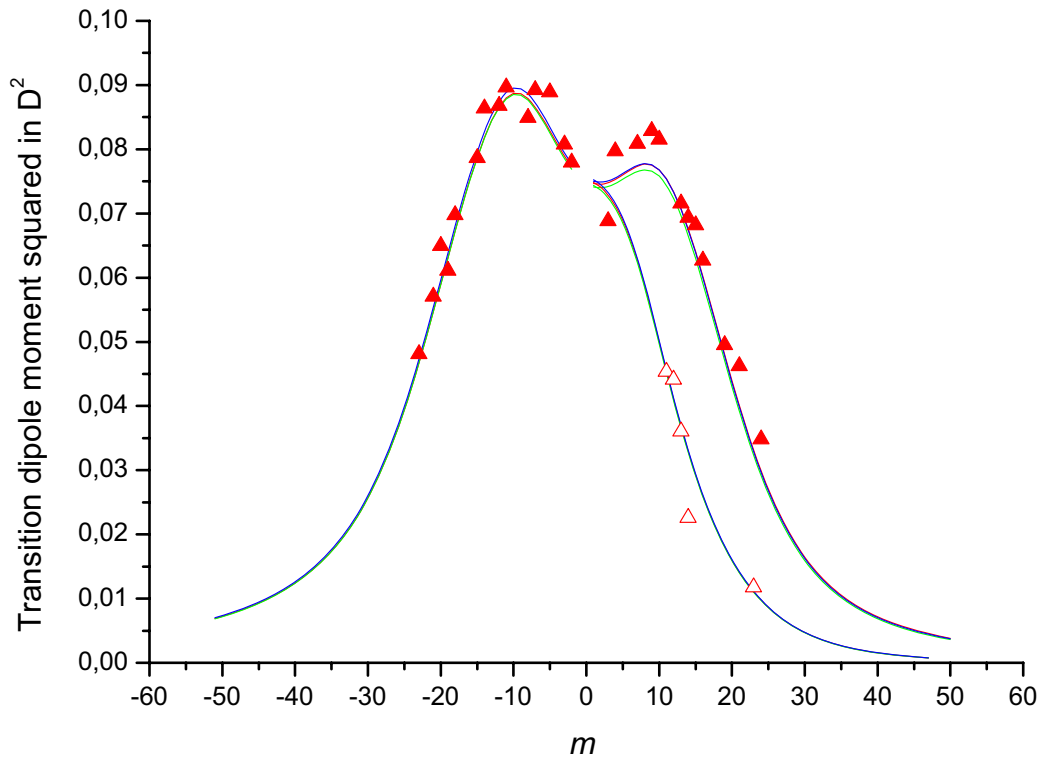


Fig. 4. Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches of the 00021 0 1 – 00020 0 0 band plotted versus  $m$  ( $m = -J, J, J+1$  for  $P, Q, R$  branch respectively) Red triangles are measurements from this work for the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches (open triangles for  $Q$ -branch and solid ones for  $P$ -and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4).

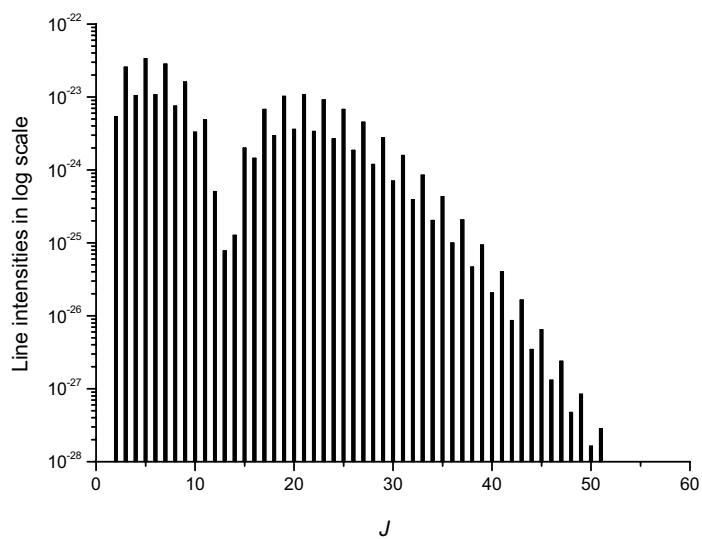


Fig. 5. Calculated line intensities (in  $\text{cm}^{-1}/(\text{molecule}\cdot\text{cm}^{-2})$  at 296K) of transitions belonging to the  $Q_{fe}$  branch of the 00031 1 1 – 00030 1 0 band plotted versus  $J$ .

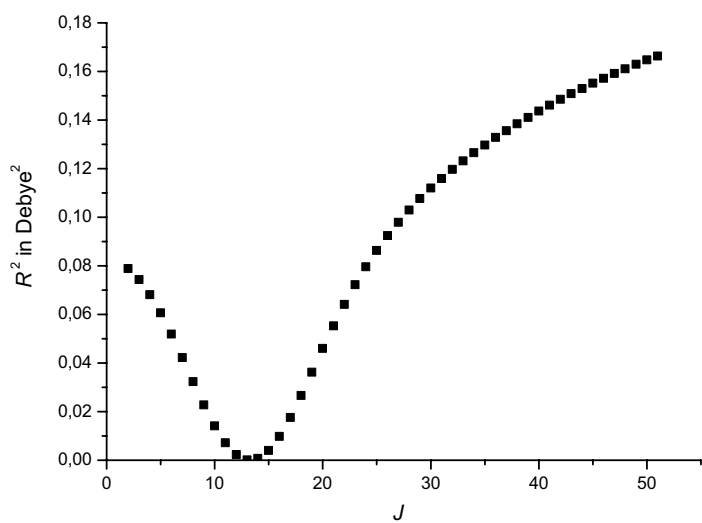


Fig. 6. Calculated transition dipole moment squared of transitions belonging to the  $Q_{fe}$  branch of the 00031 1 1 – 00030 1 0 band plotted versus  $J$ .

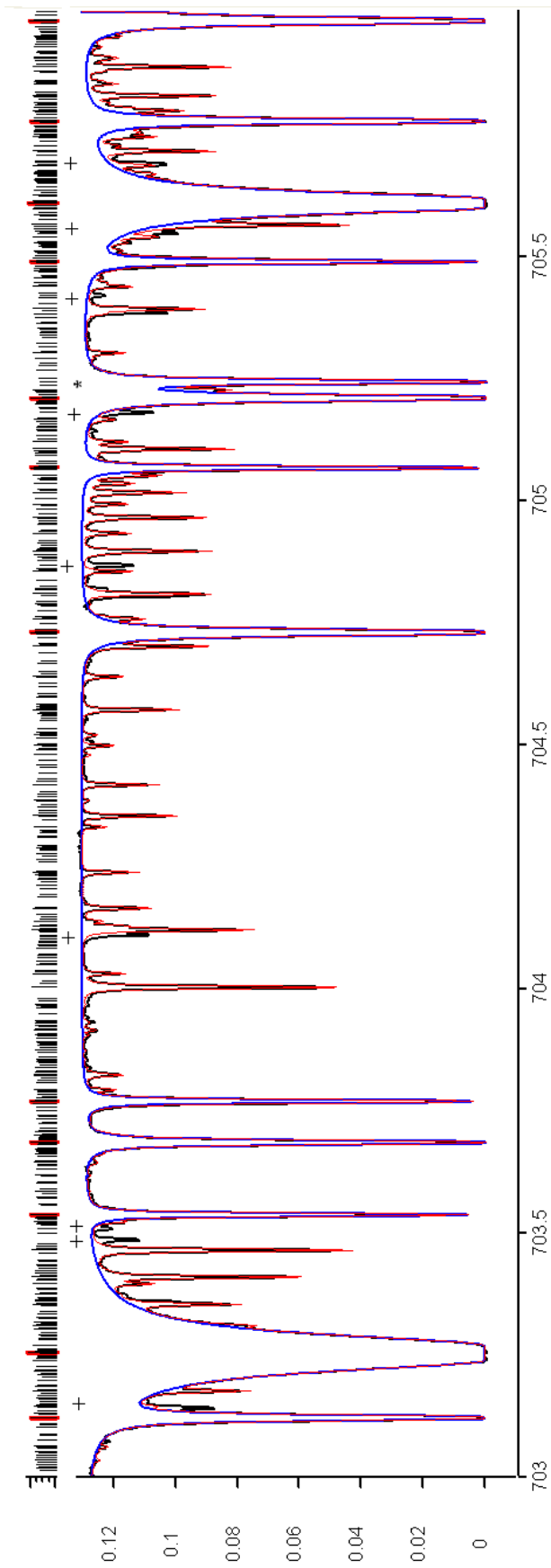


Fig. 7. Synthetic spectra between 703 and 706  $\text{cm}^{-1}$  using HITRAN 2016 or line list of this work. On the top in the first panel are plotted the transitions of  $^{12}\text{C}_2\text{H}_2$  from HITRAN2016 (red stick) and from the recommended line list of this work (black stick). In the main panel, experimental spectrum recorded at 9.3 matm of natural  $\text{C}_2\text{H}_2$  is plotted in black. Synthetic spectra are plotted in blue (HITRAN 2016) and red (recommended line list + HITRAN2016 for  $^{12}\text{C}^{13}\text{C}_2\text{H}_2$  transitions). The star symbol corresponds to the only  $^{12}\text{C}^{13}\text{C}_2\text{H}_2$  transition present in HITRAN 2016 in the plotted spectral range. The + symbols correspond to missing  $^{12}\text{C}^{13}\text{C}_2\text{H}_2$  transitions assigned from the work of Di Lonardo et al. [25].



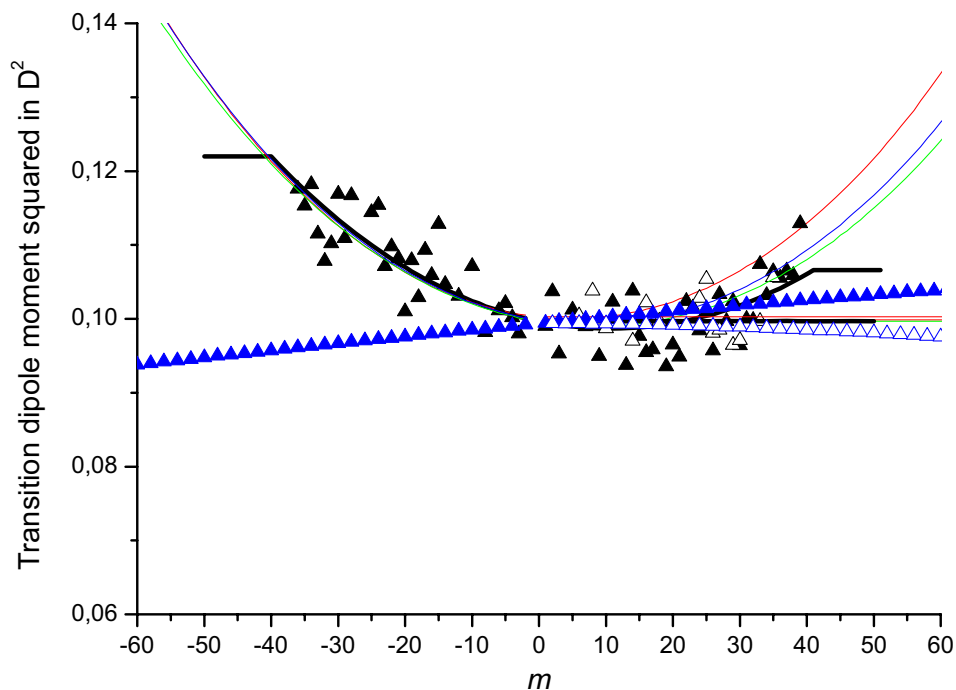


Fig. 8. Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches of the  $00001\ 0\ 1 - 00000\ 0\ 0$  band ( $\nu_5^1$ ) plotted versus  $m$  ( $m = -J, J, J+1$  for  $P, Q, R$  branch respectively). Black line is from HITRAN [11]. Black triangles are measurements from Ref. [14] (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

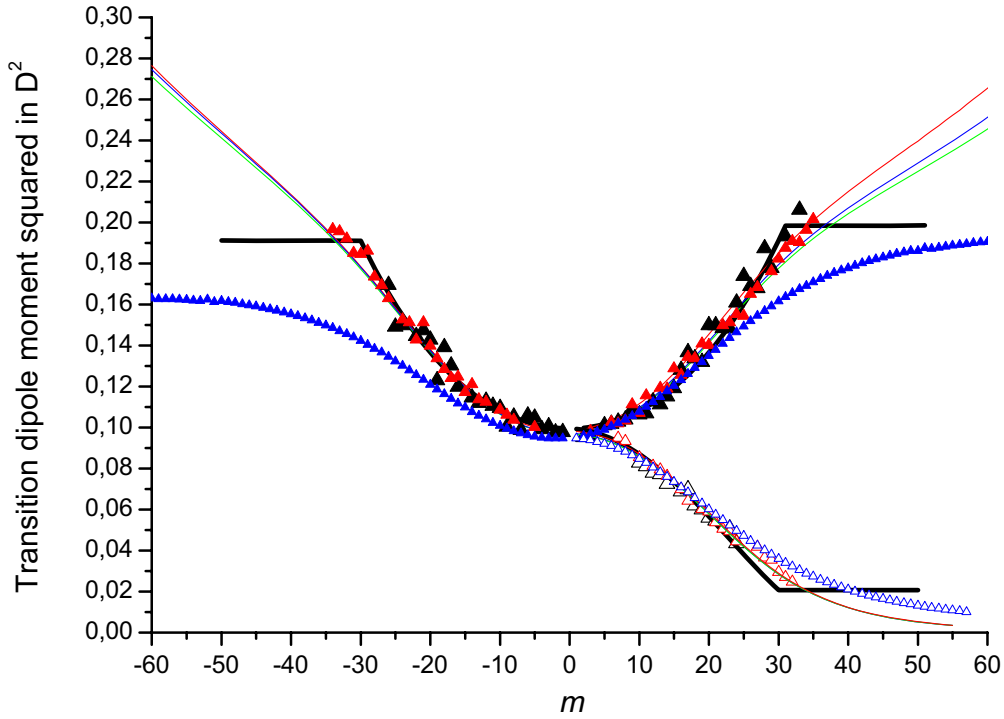


Fig. 9. Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches of the  $00002\ 0\ 0 - 00001\ 0\ 1$  band ( $2\nu_5^0 - \nu_5^1$ ) plotted versus  $m$  ( $m = -J, J, J+1$  for  $P, Q, R$  branch respectively). Black line is from HITRAN [11]. Black and red triangles are measurements from Ref. [13] and the present work respectively (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

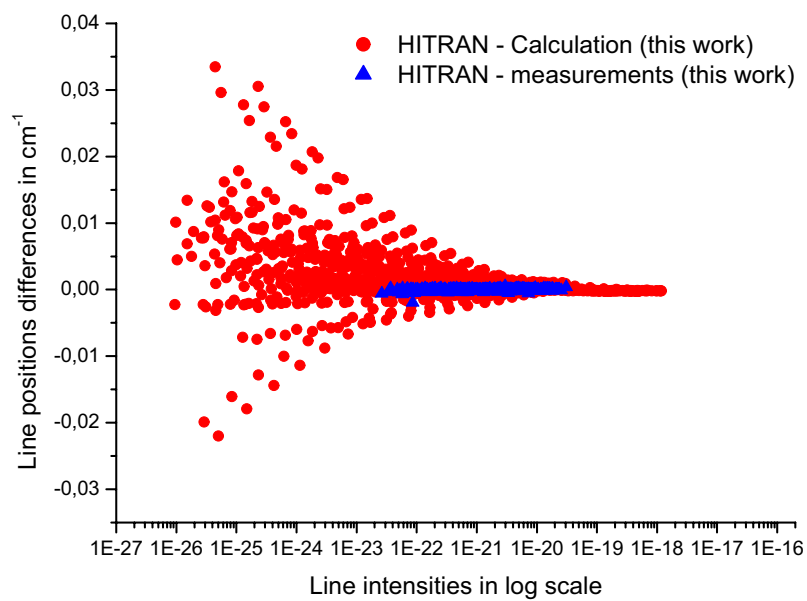


Fig. 10. Line positions differences (HITRAN – Calculation (this work)) in red circles and (HITRAN – measurements (this work)) in blue triangles plotted versus line intensities (in  $\text{cm}^{-1}/(\text{molecule}\cdot\text{cm}^{-2})$  at 296K) in a log-scale.

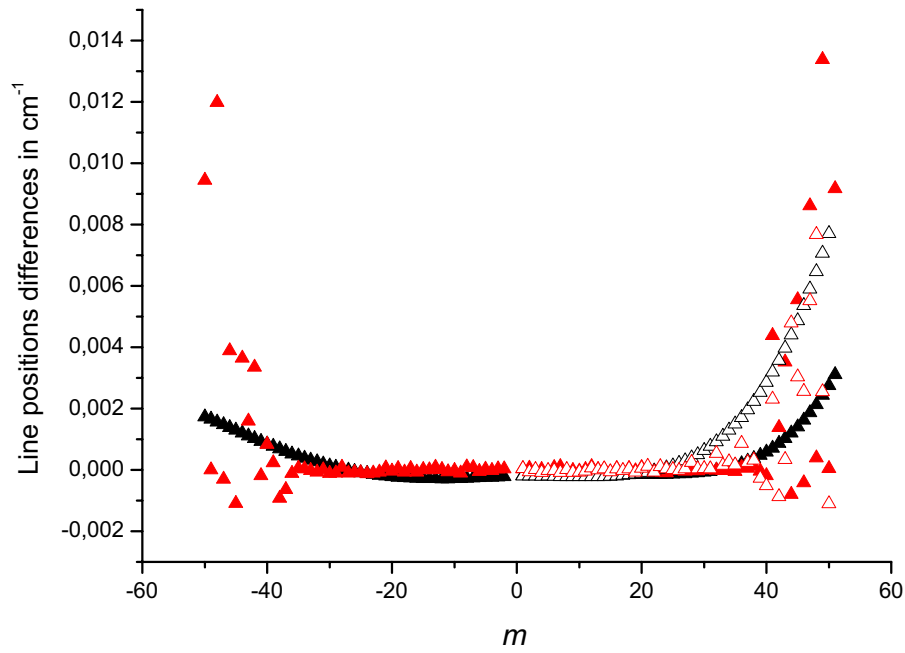


Fig. 11. Line positions differences plotted versus  $m$  ( $m = -J, J, J+1$  for  $P, Q, R$  branch respectively) for the  $00001\ 0\ 1 - 00000\ 0\ 0$  band ( $v_3^1$ ). (HITRAN - Calculation (this work)) and (HITRAN - EXOMOL) are plotted in black and red symbols respectively. Solid up-triangles correspond to  $P_{ee}$  and  $R_{ee}$ -branches, open up-triangles for  $Q_{ie}$ -branch.

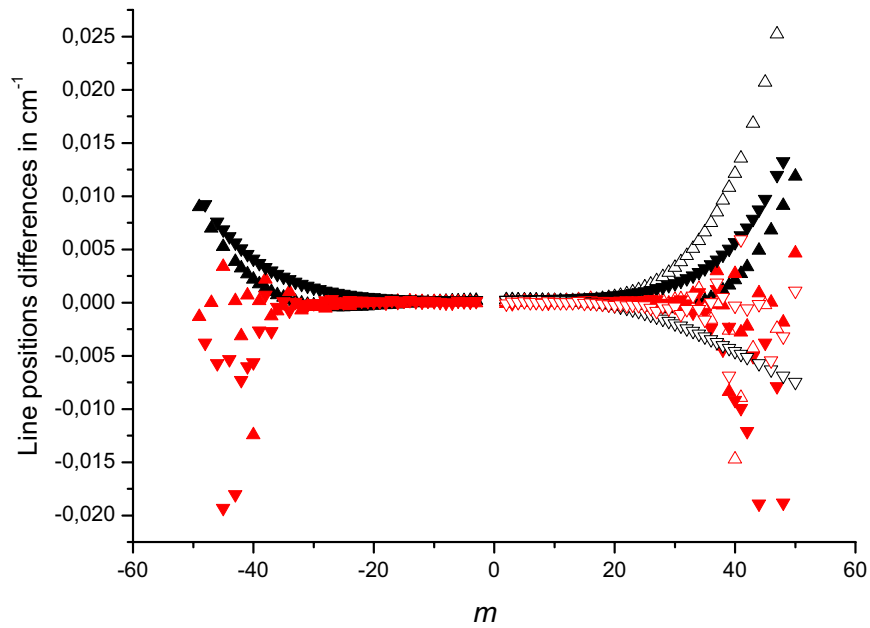


Fig. 12. Line positions differences plotted versus  $m$  ( $m = -J, J, J+1$  for  $P, Q, R$  branch respectively) for the  $00011\ 1\ 1 - 00010\ 1\ 0$  band. (HITRAN – Calculation (this work)) and (HITRAN – EXOMOL) are plotted in black and red symbols respectively. Solid up-triangles correspond to  $P_{ee}$  and  $R_{ee}$ -branches, solid down-triangles to  $P_{ff}$  and  $R_{ff}$ -branches, and open up- and down-triangles to  $Q_{fe-}$  and  $Q_{et}$ -branches respectively.

Table 1: Experimental conditions and characteristics of the recorded spectra.

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Commercial sample (Air Liquide Alphagaz)

Natural C <sub>2</sub> H <sub>2</sub>	97.760% of <sup>12</sup> C <sub>2</sub> H <sub>2</sub>
Stated purity	99.55%
Total absorption path	25.7(1) cm
Collimator focal length	418 mm
Radius of the iris	1.0 mm

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Spectrum number	Total pressure (matm)	Temperature (K)	Bruker-resolution <sup>a</sup> (cm <sup>-1</sup> )	Number of co-added scans
1	2.396	294.0	0.004	598
2	4.916	294.5	0.004	600
3	9.319	294.3	0.004	600

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<sup>a</sup> Bruker-resolution =  $0.9/\Delta_{max} \cdot \Delta_{max}$  being the maximum optical path difference.

Table 2. Measurements of line positions ( $\sigma_{\text{Obs}}$  in  $\text{cm}^{-1}$ ) and intensities ( $S_{\text{Obs}}$  in  $\text{cm}^{-1}/(\text{molecule}\cdot\text{cm}^{-2})$ ) at 296K in natural abundances) for the  $2\nu_5^0 - \nu_5^1$  band.

Assignment						$\sigma_{\text{Obs}}$	$S_{\text{Obs}}$	$m$	$R^2_{\text{Obs}}$	$E_{\text{low}}$
Pee	5	00002	0	0-00001	0 1	708.28858	0.495E-20	-5	0.1002	764.4459
Pee	8	00002	0	0-00001	0 1	701.39660	0.179E-19	-8	0.1036	813.8495
Pee	9	00002	0	0-00001	0 1	699.11600	0.611E-20	-9	0.1062	835.0207
Pee	10	00002	0	0-00001	0 1	696.84314	0.183E-19	-10	0.1086	858.5430
Pee	12	00002	0	0-00001	0 1	692.31842	0.171E-19	-12	0.1125	912.6394
Pee	13	00002	0	0-00001	0 1	690.06549	0.534E-20	-13	0.1137	943.2125
Pee	14	00002	0	0-00001	0 1	687.81789	0.155E-19	-14	0.1211	976.1350
Pee	15	00002	0	0-00001	0 1	685.57511	0.448E-20	-15	0.1172	1011.4062
Pee	16	00002	0	0-00001	0 1	683.33634	0.126E-19	-16	0.1247	1049.0256
Pee	17	00002	0	0-00001	0 1	681.10089	0.363E-20	-17	0.1241	1088.9926
Pee	18	00002	0	0-00001	0 1	678.86797	0.967E-20	-18	0.1285	1131.3064
Pee	19	00002	0	0-00001	0 1	676.63694	0.283E-20	-19	0.1336	1175.9664
Pee	20	00002	0	0-00001	0 1	674.40696	0.739E-20	-20	0.1398	1222.9719
Pee	21	00002	0	0-00001	0 1	672.17749	0.219E-20	-21	0.1512	1272.3220
Pee	22	00002	0	0-00001	0 1	669.94754	0.502E-20	-22	0.1428	1324.0159
Pee	23	00002	0	0-00001	0 1	667.71685	0.142E-20	-23	0.1512	1378.0529
Pee	24	00002	0	0-00001	0 1	665.48463	0.339E-20	-24	0.1527	1434.4319
Pee	26	00002	0	0-00001	0 1	661.01393	0.216E-20	-26	0.1630	1554.2123
Pee	27	00002	0	0-00001	0 1	658.77469	0.569E-21	-27	0.1693	1617.6118
Pee	28	00002	0	0-00001	0 1	656.53234	0.131E-20	-28	0.1735	1683.3492
Pee	29	00002	0	0-00001	0 1	654.28685	0.347E-21	-29	0.1862	1751.4240
Pee	30	00002	0	0-00001	0 1	652.03790	0.754E-21	-30	0.1846	1821.8344
Pee	31	00002	0	0-00001	0 1	649.78546	0.182E-21	-31	0.1851	1894.5793
Pee	32	00002	0	0-00001	0 1	647.52967	0.404E-21	-32	0.1921	1969.6580
Pee	33	00002	0	0-00001	0 1	645.27053	0.967E-22	-33	0.1957	2047.0688
Pee	34	00002	0	0-00001	0 1	643.00773	0.203E-21	-34	0.1967	2126.8105
Qef	7	00002	0	0-00001	0 1	719.95309	0.308E-19	7	0.09491	795.2927
Qef	8	00002	0	0-00001	0 1	719.94919	0.105E-19	8	0.09340	814.1874
Qef	10	00002	0	0-00001	0 1	719.93421	0.949E-20	10	0.08541	859.0591
Qef	12	00002	0	0-00001	0 1	719.90662	0.826E-20	12	0.08126	913.3709
Qef	13	00002	0	0-00001	0 1	719.88652	0.225E-19	13	0.07947	944.0658
Qef	14	00002	0	0-00001	0 1	719.86137	0.659E-20	14	0.07624	977.1193
Qef	16	00002	0	0-00001	0 1	719.79273	0.481E-20	16	0.06974	1050.3000
Qef	17	00002	0	0-00001	0 1	719.74779	0.116E-19	17	0.06429	1090.4258
Qef	19	00002	0	0-00001	0 1	719.63261	0.792E-20	19	0.06020	1177.7453
Qef	20	00002	0	0-00001	0 1	719.56104	0.216E-20	20	0.05887	1224.9374
Qef	21	00002	0	0-00001	0 1	719.47910	0.487E-20	21	0.05376	1274.4834
Qef	22	00002	0	0-00001	0 1	719.38625	0.124E-20	22	0.05060	1326.3824
Qef	23	00002	0	0-00001	0 1	719.28190	0.291E-20	23	0.04927	1380.6335
Qef	24	00002	0	0-00001	0 1	719.16548	0.691E-21	24	0.04436	1437.2359
Qef	27	00002	0	0-00001	0 1	718.73992	0.897E-21	27	0.04182	1621.1406
Qef	28	00002	0	0-00001	0 1	718.57184	0.196E-21	28	0.03653	1687.1379
Qef	29	00002	0	0-00001	0 1	718.38992	0.424E-21	29	0.03537	1755.4812
Qef	30	00002	0	0-00001	0 1	718.19631	0.864E-22	30	0.02954	1826.1693
Qef	31	00002	0	0-00001	0 1	717.98538	0.171E-21	31	0.02695	1899.2012
Qef	32	00002	0	0-00001	0 1	717.76295	0.374E-22	32	0.02467	1974.5755
Ree	2	00002	0	0-00001	0 1	727.07335	0.573E-20	3	0.09818	736.2127
Ree	5	00002	0	0-00001	0 1	734.26923	0.440E-20	6	0.01026	764.4459

Ree 6	00002	0	0-00001	0	1	736.68432	0.147E-19	7	0.01020	778.5618
Ree 7	00002	0	0-00001	0	1	739.10696	0.538E-20	8	0.01034	795.0298
Ree 8	00002	0	0-00001	0	1	741.53660	0.182E-19	9	0.01113	813.8495
Ree 9	00002	0	0-00001	0	1	743.97281	0.595E-20	10	0.01071	835.0207
Ree 10	00002	0	0-00001	0	1	746.41480	0.191E-19	11	0.01156	858.5430
Ree 12	00002	0	0-00001	0	1	751.31350	0.184E-19	13	0.01193	912.6394
Ree 13	00002	0	0-00001	0	1	753.76882	0.571E-20	14	0.01189	943.2125
Ree 14	00002	0	0-00001	0	1	756.22700	0.171E-19	15	0.01287	976.1350
Ree 15	00002	0	0-00001	0	1	758.68726	0.504E-20	16	0.01256	1011.4062
Ree 16	00002	0	0-00001	0	1	761.14881	0.144E-19	17	0.01344	1049.0256
Ree 17	00002	0	0-00001	0	1	763.61084	0.420E-20	18	0.01337	1088.9926
Ree 18	00002	0	0-00001	0	1	766.07248	0.115E-19	19	0.01409	1131.3064
Ree 19	00002	0	0-00001	0	1	768.53302	0.324E-20	20	0.01401	1175.9664
Ree 21	00002	0	0-00001	0	1	773.44775	0.242E-20	22	0.01499	1272.3220
Ree 22	00002	0	0-00001	0	1	775.90062	0.598E-20	23	0.01512	1324.0159
Ree 23	00002	0	0-00001	0	1	778.34968	0.165E-20	24	0.01553	1378.0529
Ree 24	00002	0	0-00001	0	1	780.79446	0.392E-20	25	0.01543	1434.4319
Ree 25	00002	0	0-00001	0	1	783.23451	0.110E-20	26	0.01651	1493.1520
Ree 26	00002	0	0-00001	0	1	785.66942	0.261E-20	27	0.01685	1554.2123
Ree 28	00002	0	0-00001	0	1	790.52292	0.158E-20	29	0.01761	1683.3492
Ree 29	00002	0	0-00001	0	1	792.94117	0.406E-21	30	0.01823	1751.4240
Ree 30	00002	0	0-00001	0	1	795.35350	0.924E-21	31	0.01875	1821.8344
Ree 31	00002	0	0-00001	0	1	797.76000	0.228E-21	32	0.01909	1894.5793
Ree 32	00002	0	0-00001	0	1	800.16051	0.491E-21	33	0.01903	1969.6580
Ree 33	00002	0	0-00001	0	1	802.55513	0.120E-21	34	0.01963	2047.0688
Ree 34	00002	0	0-00001	0	1	804.94402	0.259E-21	35	0.02016	2126.8105

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Note: For assignment, the branch type is first given, then the *ef* Wang symmetry of the upper and lower state, and finally the rotational quantum number *J* of the lower state. Then are reported the vibrational assignment for the upper and lower states ( $V_1 V_2 V_3 V_4 V_5 \ell_4 \ell_5$ ). Measured line positions ( $\sigma_{\text{Meas}}$ ) and energies of the lower state ( $E_{\text{low}}$ ) are given in  $\text{cm}^{-1}$ . The transition dipole moments squared ( $R^2_{\text{Obs}}$ ) are given in  $\text{debye}^2$ .



Table 3. Summary of measurements performed in this work.

Studied band	$\Delta\ell_4 \Delta\ell_5 N$			Positions min - max	Line intensities min - max	$J$ min - max
00002 0 0 *	00001 0 1	0 -1	74	643.0 - 804.9	3.74E-23 - 3.08E-20	2 - 34
00002 0 2 *	00001 0 1	0 -1	90	651.0 - 819.6	3.55E-23 - 2.36E-20	2 - 37
00011 1-1 *	00010 1 0	0 1	122	638.3 - 806.0	3.70E-23 - 2.55E-20	1 - 35
00011 1 1 *	00010 1 0	0 1	89	657.5 - 812.6	2.64E-23 - 2.31E-20	4 - 34
00012 1 0	00011 1-1	0 1	108	638.3 - 805.3	1.03E-23 - 2.52E-21	0 - 34
00012 1 2	00011 1 1	0 1	60	673.8 - 795.8	4.54E-23 - 1.57E-21	6 - 26
00003 0 1	00002 0 0	0 1	51	660.2 - 784.7	4.39E-23 - 1.75E-21	1 - 29
00003 0 3	00002 0 2	0 1	46	675.3 - 789.7	6.25E-23 - 1.29E-21	6 - 24
00021 0 1	00020 0 0	0 1	31	674.4 - 785.2	6.11E-23 - 1.47E-21	2 - 23
00021 0 1	00020 2 0	-2 1	58	676.3 - 802.1	1.15E-23 - 1.26E-21	2 - 28
00021 2 1	00020 2 0	0 1	81	674.0 - 809.8	1.04E-23 - 1.41E-21	4 - 30
00021 2-1	00020 2 0	0 -1	108	649.5 - 794.5	1.24E-23 - 1.00E-21	2 - 32
00021 2-1	00020 0 0	2 -1	32	673.2 - 754.7	1.11E-23 - 1.16E-21	0 - 27
00012 1 0	00011 1 1	0 -1	47	654.5 - 765.9	4.25E-23 - 4.36E-22	2 - 23
00012-1 2	00011 1-1	-2 3	59	660.8 - 789.3	1.02E-23 - 1.20E-22	1 - 24
00012-1 2	00011 1 1	-2 1	59	668.9 - 779.7	2.71E-23 - 3.92E-22	3 - 26
00003 0 1	00002 0 2	0 -1	68	649.4 - 779.4	1.02E-23 - 5.00E-22	2 - 29
00021 2 1	00020 0 0	2 1	37	662.1 - 815.2	1.15E-23 - 4.63E-22	3 - 35

Note: The band notation refers to the upper and lower vibrational state, see note of Table 2. The line positions are given in  $\text{cm}^{-1}$  and the line intensities are in  $\text{cm}^{-1}/(\text{molecule}\cdot\text{cm}^{-2})$  at 296K in natural abundance. The star marks the bands already measured in Ref. [13] and present in HITRAN and GEISA databases.

Table 4. Effective dipole moment parameters for the  $\Delta P=1$  series of transitions.

Parameter <sup>a</sup>	$\Delta V_1$	$\Delta V_2$	$\Delta V_3$	$\Delta V_4$	$\Delta V_5$	$\Delta \ell_4$	$\Delta \ell_5$	<i>This work</i>	Ref. [1]	Ref. [4]
$M$	0	0	0	0	1	0	1	-0.1583(2)	-0.1578(2)	-0.1579(2)
$\kappa_5$	0	0	0	0	1	0	1	-0.0131(8)	-0.017(2)	0.013(2)
$b_J$	0	0	0	0	1	0	1	$-0.49(3) \times 10^{-3}$	$-0.76(5) \times 10^{-3}$	$-0.72(5) \times 10^{-3}$
$d_J$	0	0	0	0	1	0	1	$0.51(2) \times 10^{-4}$	$0.45(3) \times 10^{-4}$	$0.47(3) \times 10^{-4}$
$M$	0	0	0	2	-1	2	-1	$-0.3361(4) \times 10^{-2}$	$-0.3343(5) \times 10^{-2}$	
$\kappa_4$	0	0	0	2	-1	2	-1	0.028(4)	0.020(3)	
$b_J$	0	0	0	2	-1	2	-1	$-0.211(8) \times 10^{-2}$	$-0.244(9) \times 10^{-2}$	
$d_J$	0	0	0	2	-1	2	-1	$-0.66(5) \times 10^{-4}$	$-0.63(6) \times 10^{-4}$	
$M$	0	0	0	2	-1	0	1	$0.669(2) \times 10^{-2}$	$0.665(2) \times 10^{-2}$	
$\kappa_4$	0	0	0	2	-1	0	1	0.060(2)	0.059(2)	
$a_5$	0	0	0	2	-1	0	1	0.030(2)	0.026(2)	
$b_J$	0	0	0	2	-1	0	1	$-0.109(7) \times 10^{-2}$	$-0.119(8) \times 10^{-2}$	
$d_J$	0	0	0	2	-1	0	1	$-0.55(4) \times 10^{-4}$	$-0.61(4) \times 10^{-4}$	
$M$	0	0	0	2	-1	-2	3	$-0.36(3) \times 10^{-3}$	$-0.30(3) \times 10^{-3}$	

<sup>a</sup> Parameters  $M$  are given in Debye while the other parameters are dimensionless.

<sup>b</sup> Confidence intervals (1 SD, in unit of the last quoted digit) are given between parentheses.

Table 5. Statistics of the fitted bands of the  $\Delta P = 1$  series.

Measured bands	Spectral range in $\text{cm}^{-1}$	$N^a$	$J_{\max}^b$	MR <sup>c</sup> in %	RMS <sup>d</sup> in %
<b>Band measured in Ref. [14]</b>					
00001 0 1 - 00000 0 0	644.5-820.2	77	38	-1.10	4.01
<b>Bands measured in Ref. [13]</b>					
00002 0 0 - 00001 0 1	661.0-800.2	64	32	-1.13	3.79
00002 0 2 - 00001 0 1	674.7-794.8	79	30	-1.01	2.67
00011 1-1 - 00010 1 0	655.8-787.5	109	31	-0.13	3.31
00011 1 1 - 00010 1 0	669.4-798.8	98	28	-0.15	4.19
<b>Bands measured in Ref. [1]</b>					
00020 2 0 - 00001 0 1	434.6-564.0	107	33	-1.08	5.82
00020 0 0 - 00001 0 1	429.6-586.8	84	34	1.77	4.72
00030 1 0 - 00011 1 1	449.7-583.9	77	27	-1.08	4.56
00030 1 0 - 00011 1-1	478.9-580.6	37	22	1.41	6.29
00030 3 0 - 00011 1 1	468.3-559.8	40	25	0.85	5.54
00021 0 1 - 00002 0 0	457.4-568.3	33	23	0.93	5.80
00021 0 1 - 00002 0 2	458.6-527.2	52	25	-0.11	3.45
00021 2 1 - 00002 0 2	456.8-555.6	20	21	4.73	8.88
00021 2 1 - 00002 0 0	555.7-565.6	2	21	6.89	6.92
<b>Bands measured in this work</b>					
00002 0 0 - 00001 0 1	643.0-804.9	72	34	-0.13	2.69
00002 0 2 - 00001 0 1	651.0-819.6	87	37	0.48	3.33
00011 1-1 - 00010 1 0	638.3-806.0	119	35	0.27	3.54
00011 1 1 - 00010 1 0	657.5-812.6	85	34	-0.51	3.74
00003 0 1 - 00002 0 0	660.2-784.7	47	29	1.49	3.99
00003 0 1 - 00002 0 2	649.4-779.4	64	29	-1.20	4.48
00003 0 3 - 00002 0 2	675.3-784.7	40	22	-0.95	3.59
00012 1 0 - 00011 1 1	654.5-765.9	46	23	-0.96	4.53
00012 1 0 - 00011 1-1	638.3-795.6	65	34	1.92	4.99
00012 1 2 - 00011 1 1	673.8-795.8	50	26	-0.15	4.46
00012-1 2 - 00011 1 1	668.9-779.7	54	26	2.27	4.29
00012-1 2 - 00011 1-1	652.3-805.3	48	33	3.29	6.72
00021 0 1 - 00020 0 0	674.3-774.0	28	23	3.49	4.72
00021 0 1 - 00020 2 0	676.3-802.1	28	28	6.32	8.16
00021 2 1 - 00020 0 0	662.1-810.6	22	33	0.95	6.06
00021 2 1 - 00020 2 0	674.0-809.8	64	30	0.30	3.86
00021 2-1 - 00020 0 0	673.2-754.7	26	27	-1.42	3.80
00021 2-1 - 00020 2 0	649.5-794.5	99	32	-0.19	3.72

Note: The band notation refers to the upper and lower vibrational states ( $V_1 V_2 V_3 V_4 V_5 \ell_4 \ell_5$ ).

<sup>a</sup> Number of the fitted line intensities for a given band.

<sup>b</sup> Maximum value of the angular momentum quantum number for a given band.

<sup>c</sup> Mean residuals.

<sup>d</sup> Root mean squares of the residuals.

Table 6. Calculation based on parameter of Table 4 (this work) for the  $Q_{fe}$  and  $R_{ce}$  branches of the 00031 1 1 – 00030 1 0 band. The first column corresponds to the vibrational labeling used for a whole series of transition. Then the type of branch, the Wang symmetry  $e/f$  of the upper and lower state,  $J$  of lower state, calculated line positions in  $\text{cm}^{-1}$ , calculated line intensities at 296K for natural abundances in  $\text{cm}^{-1}/(\text{molecule}\cdot\text{cm}^{-2})$ , the lower state energy  $\text{cm}^{-1}$ , then, for upper and lower levels, the two main contributors to the eigenfunctions (with percentages, see text). A star at the beginning of the line means that the main contributors to the eigenfunctions are different from the vibrational labeling used for a whole series of transition.

Vibrational labeling of the band		Br	$J$	Position	Intensity	$E_{Low}$	Upper main contributors		Lower main contributors																	
							First	per	Second	per	First	per	Second	per												
00031	1	1-00030	1	0	Qfe	14	731.74096	1.272E-25	2101.70012	00031	1	1	46%	00031	1-1	26%	00030	1	0	88%	00030	3	0	11%		
00031	1	1-00030	1	0	Qfe	15	731.87770	2.005E-24	2137.05022	00031	1	1	39%	00031	1-1	28%	00030	1	0	84%	00030	3	0	14%		
00031	1	1-00030	1	0	Qfe	16	732.00377	1.449E-24	2174.76900	00031	1	1	33%	00031	1-1	31%	00030	1	0	81%	00030	3	0	18%		
*	00031	1	1-00030	1	0	Qfe	17	732.11764	2214.85691	00031	1-1	1	32%	00031	3	1	30%	00030	1	0	77%	00030	3	0	22%	
*	00031	1	1-00030	1	0	Qfe	18	732.21895	2.949E-24	2257.31374	00031	3	1	35%	00031	1-1	34%	00030	1	0	73%	00030	3	0	26%	
*	00031	1	1-00030	1	0	Qfe	19	732.30829	1.022E-23	2302.13864	00031	3	1	39%	00031	1-1	35%	00030	1	0	69%	00030	3	0	30%	
*	00031	1	1-00030	1	0	Qfe	20	732.38697	3.613E-24	2349.33035	00031	3	1	42%	00031	1-1	35%	00030	1	0	65%	00030	3	0	34%	
*	00031	1	1-00030	1	0	Qfe	21	732.45667	1.076E-23	2398.88729	00031	3	1	45%	00031	1-1	36%	00030	1	0	62%	00030	3	0	37%	
*	00031	1	1-00030	1	0	Qfe	22	732.51925	3.381E-24	2450.80772	00031	3	1	48%	00031	1-1	36%	00030	1	0	59%	00030	3	0	40%	
*	00031	1	1-00030	1	0	Qfe	23	732.57660	9.170E-24	2505.08983	00031	3	1	50%	00031	1-1	36%	00030	1	0	56%	00030	3	0	43%	
...																										
00031	1	1-00030	1	0	Ree	23	787.55801	2.763E-25	2505.08983	00031	1	1	47%	00031	3	1	39%	00030	1	0	56%	00030	3	0	43%	
00031	1	1-00030	1	0	Ree	24	789.80567	4.481E-26	2561.73183	00031	1	1	46%	00031	3	1	41%	00030	1	0	53%	00030	3	0	46%	
00031	1	1-00030	1	0	Ree	25	792.04317	5.871E-26	2620.73195	00031	1	1	44%	00031	3	1	42%	00030	1	0	51%	00030	3	0	48%	
*	00031	1	1-00030	1	0	Ree	26	794.27160	7.192E-27	2682.08846	00031	3	1	44%	00031	1	1	43%	00030	3	0	50%	00030	1	0	49%
*	00031	1	1-00030	1	0	Ree	27	796.49193	5.664E-27	2745.79968	00031	3	1	45%	00031	1	1	41%	00030	3	0	52%	00030	1	0	47%
*	00031	1	1-00030	1	0	Ree	28	798.70503	1.744E-28	2811.86399	00031	3	1	46%	00031	1	1	40%	00030	3	0	53%	00030	1	0	45%
*	00031	1	1-00030	1	0	Ree	29	800.91165	2.012E-28	2880.27978	00031	3	1	47%	00031	1	1	38%	00030	3	0	55%	00030	1	0	44%
*	00031	1	1-00030	1	0	Ree	30	803.11247	5.125E-28	2951.04548	00031	3	1	48%	00031	1	1	37%	00030	3	0	56%	00030	1	0	43%
*	00031	1	1-00030	1	0	Ree	31	805.30808	3.021E-27	3024.15952	00031	3	1	49%	00031	1	1	36%	00030	3	0	57%	00030	1	0	41%
*	00031	1	1-00030	1	0	Ree	32	807.49901	1.353E-27	3099.62035	00031	3	1	50%	00031	1	1	35%	00030	3	0	58%	00030	1	0	40%
*	00031	1	1-00030	1	0	Ree	33	809.68572	4.524E-27	3177.42640	00031	3	1	51%	00031	1	1	34%	00030	3	0	59%	00030	1	0	39%

Note: The vibrational labeling  $V_1 V_2 V_3 V_4 V_5 \ell_4 \ell_5$  is used for upper and lower state.

Table 7. Summary of calculation for the 30 first strongest bands in the  $\Delta P = 1$  series of bands. The first column corresponds to the vibrational labeling used for series of transitions (bands).  $N$  is the total number of transitions belonging to band. Minimum and maximum wavenumbers of calculated transitions belonging to the band are given in  $\text{cm}^{-1}$ ,  $Sum\_S$  is the sum of calculated line intensities (see text) in  $\text{cm}^{-1}/(\text{molecule.cm}^{-2})$  at 296K for natural abundances.  $Smin$  and  $Smax$  are respectively the minimum and maximum calculated line intensities in the band. Minimum and maximum values of  $J$  in the band are given.

Vibrational labeling of the band		$N$	Wavenumbers min – max	$Sum\_S$	$Smin$	$Smax$	$J$ min max		
H	00001 0 1 _ 00000 0 0	195	577.0-881.5	2.46E-17	1.36E-28	1.18E-18	0	65	Obs
H	00002 0 2 _ 00001 0 1	343	589.8-887.6	6.94E-19	1.32E-28	3.59E-20	1	61	Obs
H	00002 0 0 _ 00001 0 1	176	583.5-869.8	6.89E-19	1.40E-28	3.01E-20	1	62	Obs
H	00011 1 1 _ 00010 1 0	345	588.7-892.7	6.33E-19	1.09E-28	3.18E-20	1	62	Obs
H	00011 1-1 _ 00010 1 0	355	579.9-866.0	6.23E-19	1.08E-28	3.11E-20	1	61	Obs
	00003 0 1 _ 00002 0 0	167	585.7-853.8	4.07E-20	1.07E-28	1.87E-21	0	57	Obs
*	00021 0 1 _ 00020 0 0	149	604.2-846.0	3.62E-20	1.13E-28	1.80E-21	0	51	Obs
	00012 1 2 _ 00011 1 1	315	603.8-871.6	3.49E-20	1.00E-28	1.63E-21	2	58	Obs
	00003 0 3 _ 00002 0 2	313	598.9-868.5	2.89E-20	1.02E-28	1.36E-21	2	57	Obs
*	00012 1 0 _ 00011 1-1	319	587.1-854.9	2.79E-20	1.01E-28	2.54E-21	0	58	Obs
*	00012-1 2 _ 00011 1-1	302	601.6-862.4	2.69E-20	1.18E-28	2.54E-21	0	57	Obs
*	00021 2 1 _ 00020 2 0	282	627.2-881.0	2.58E-20	1.01E-28	1.40E-21	2	54	Obs
*	00021 2-1 _ 00020 2 0	321	582.9-853.7	2.26E-20	1.10E-28	1.02E-21	2	57	Obs
	00021 2-1 _ 00020 0 0	126	630.2-798.0	1.49E-20	1.16E-28	1.20E-21	0	53	Obs
*	00021 0 1 _ 00020 2 0	310	612.6-879.5	1.27E-20	1.25E-28	1.46E-21	2	57	Obs
*	00021 2 1 _ 00020 0 0	162	600.1-865.0	1.05E-20	1.06E-28	4.70E-22	2	57	Obs
	00003 0 1 _ 00002 0 2	298	590.2-854.8	9.41E-21	1.02E-28	5.07E-22	2	56	Obs
*	00012-1 2 _ 00011 1 1	307	594.9-859.1	8.56E-21	1.25E-28	3.92E-22	2	57	Obs
*	00012 1 0 _ 00011 1 1	297	578.7-843.6	8.46E-21	1.01E-28	4.44E-22	2	53	Obs
	01001 0 1 _ 01000 0 0	151	608.3-847.6	1.69E-21	1.15E-28	8.11E-23	0	51	
*	00022 0 0 _ 00021 0 1	143	620.0-838.9	1.65E-21	1.19E-28	8.28E-23	1	51	
*	00022 2 2 _ 00021 2 1	277	616.6-854.1	1.64E-21	1.34E-28	9.87E-23	3	51	
	00022 2-2 _ 00021 2-1	297	590.6-839.7	1.64E-21	1.08E-28	8.05E-23	1	52	
*	00022 0 2 _ 00021 0 1	284	621.7-854.5	1.45E-21	1.05E-28	7.59E-23	1	52	
	00013 1 3 _ 00012 1 2	278	619.9-855.5	1.42E-21	1.11E-28	8.36E-23	3	52	
*	00013 1 1 _ 00012 1 0	282	598.9-855.5	1.35E-21	1.07E-28	7.41E-23	1	52	
*	00031 1-1 _ 00030 1 0	254	598.1-840.2	1.27E-21	1.05E-28	1.02E-22	1	51	
*	00013-1 3 _ 00012-1 2	280	609.4-844.8	1.27E-21	1.12E-28	6.78E-23	1	51	
	00004 0 0 _ 00003 0 1	146	597.2-837.8	1.18E-21	1.18E-28	5.00E-23	1	52	
*	00031 3-1 _ 00030 3 0	283	590.6-837.0	1.18E-21	1.05E-28	5.09E-23	3	52	

Note: The vibrational labeling for upper and lower state corresponds to  $V_1 V_2 V_3 V_4 V_5 \ell_4 \ell_5$ . A star at the beginning of the line means that the main contributor to the eigenfunctions of the upper or lower states (or both) is changed within the series of transitions (bands). Letter H shows bands present in HITRAN2016 [11]. In the last column ‘‘Obs’’ means that the band was measured in this work or reported in literature (see Section 2).

## Declaration of interests

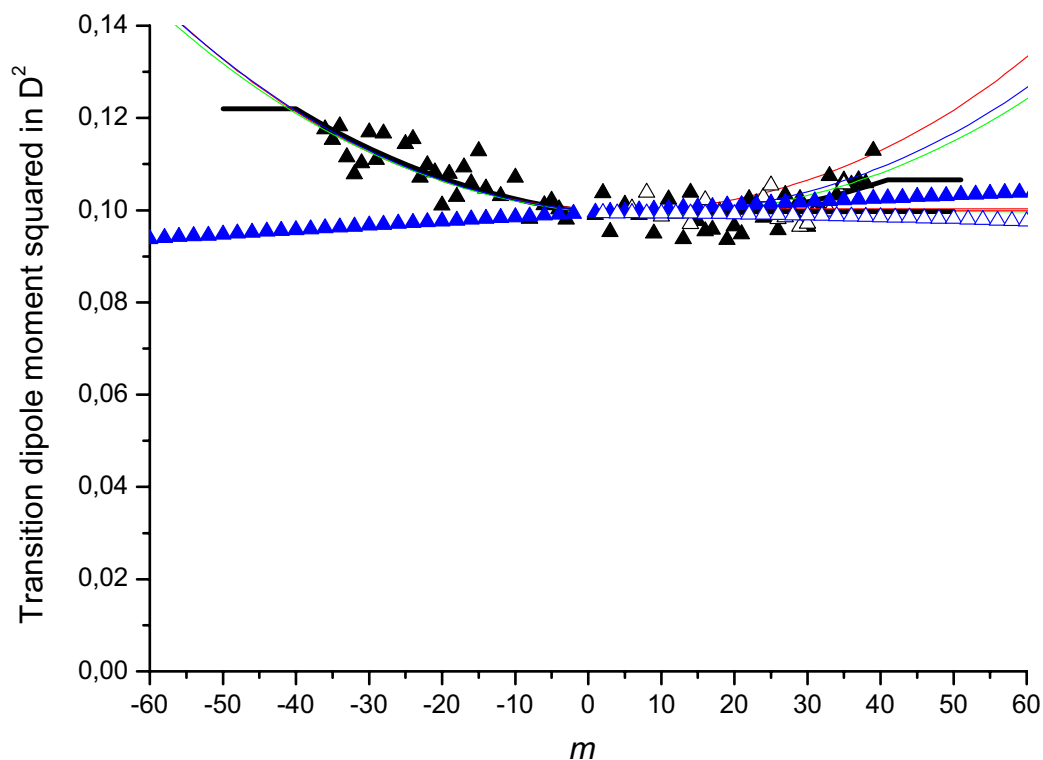
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

**David Jacquemart:** Visualization, Analysis, Investigation, Methodology, Data Curation, Writing - Original Draft. **Pascale Soulard:** Investigation. **Oleg Lyulin:** Methodology, Software, Validation, Formal analysis, Data Curation, Writing - Original Draft.

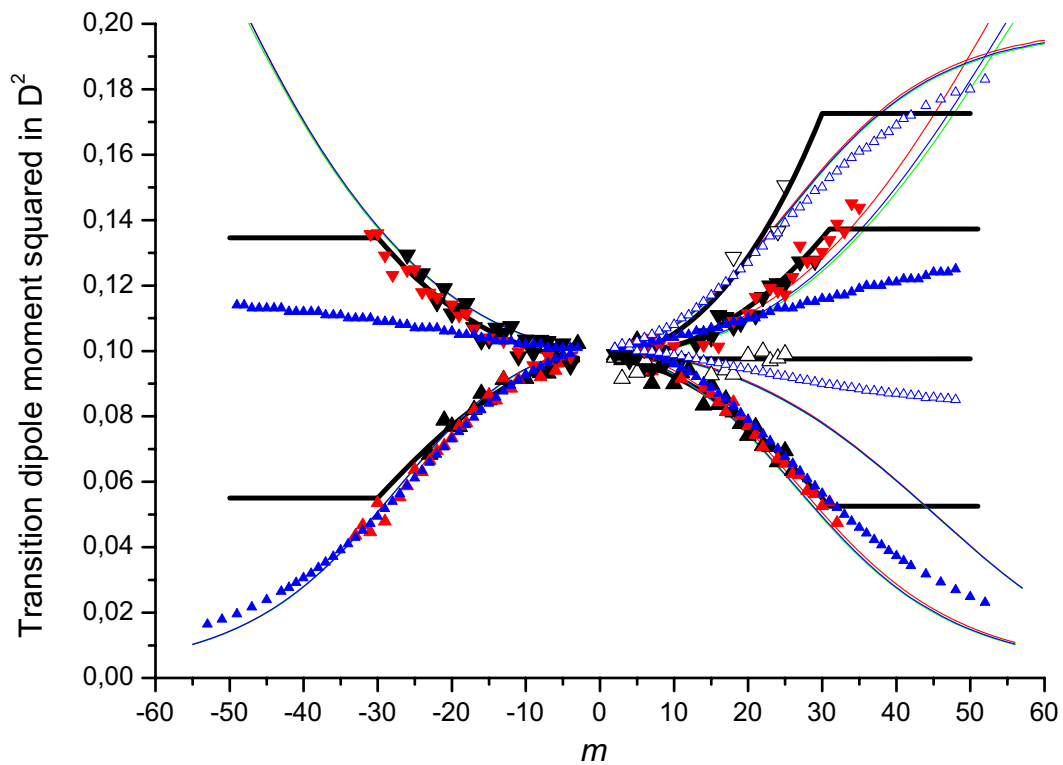
## Supplementary material 2:

Comparisons of  $R^2$  experimental values with  $R^2$  values from HITRAN/EXOMOL/Global model [1,4,present work].

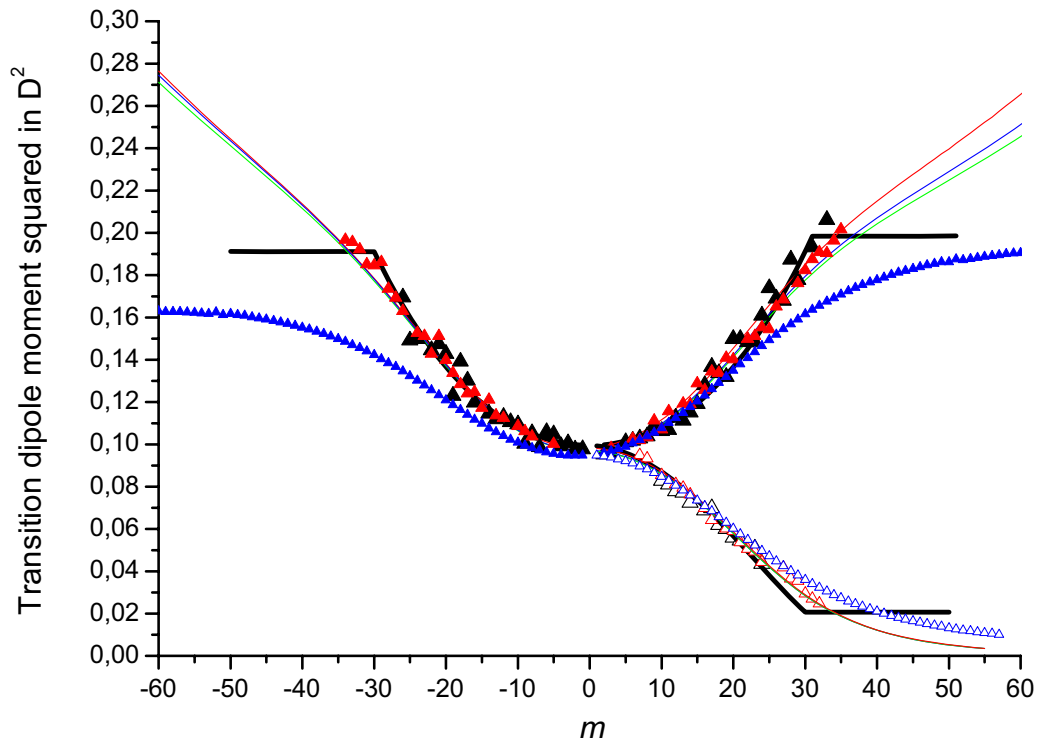


Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches of the 00001 0 1 – 00000 0 0 band ( $\nu_5^1$ ) plotted versus  $m$ . Black line is from HITRAN [11]. Black triangles are measurements from Ref. [14] (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

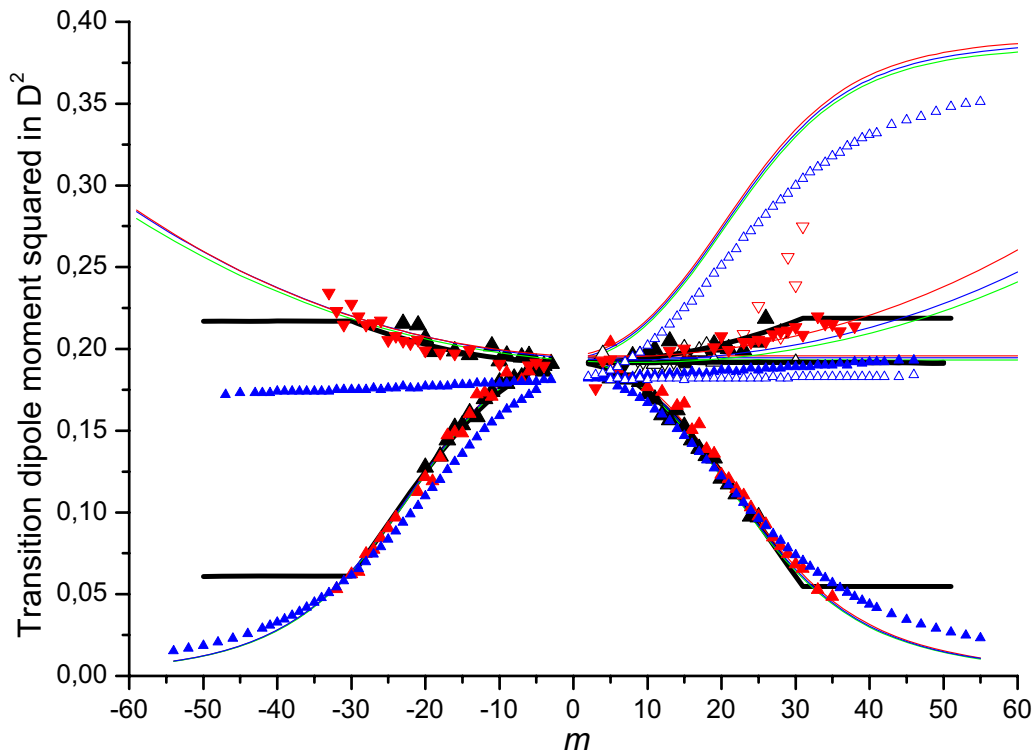




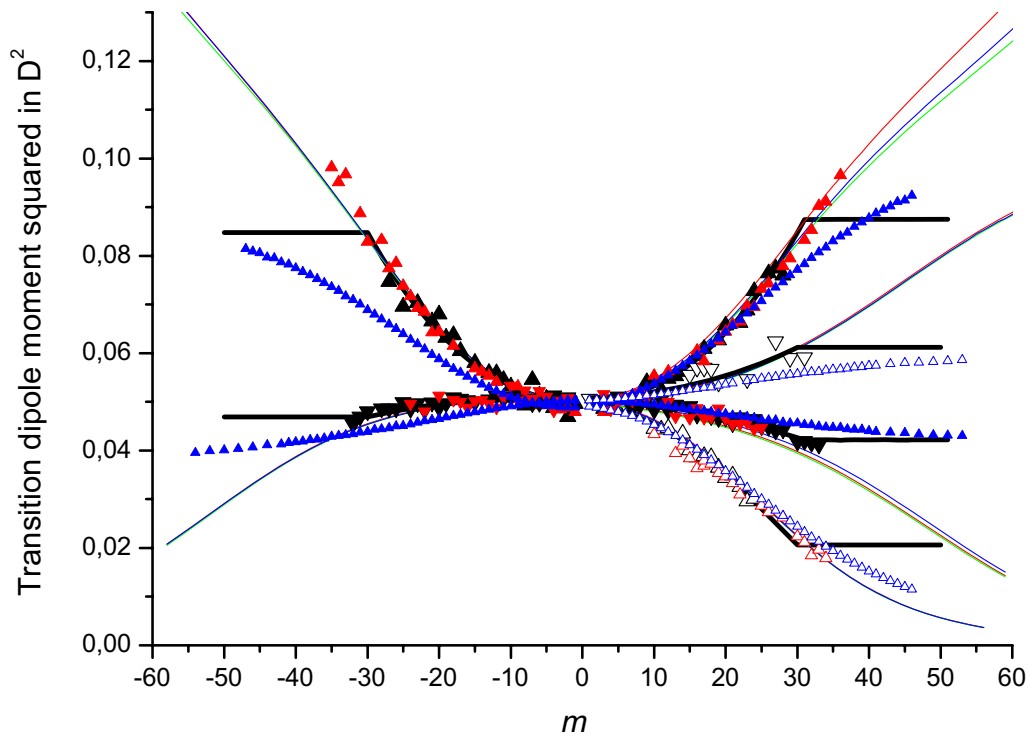
Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00011\ 1\ 1 - 00010\ 1\ 0$  band plotted versus  $m$  (noted  $(v_4+v_5)^2 - v_4^1$ ) in Ref. [13] as well as in HITRAN and GEISA databases). Black line is from HITRAN [11]. Black and red triangles are measurements from Ref. [13] and the present work respectively (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Upside triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas downside triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4).



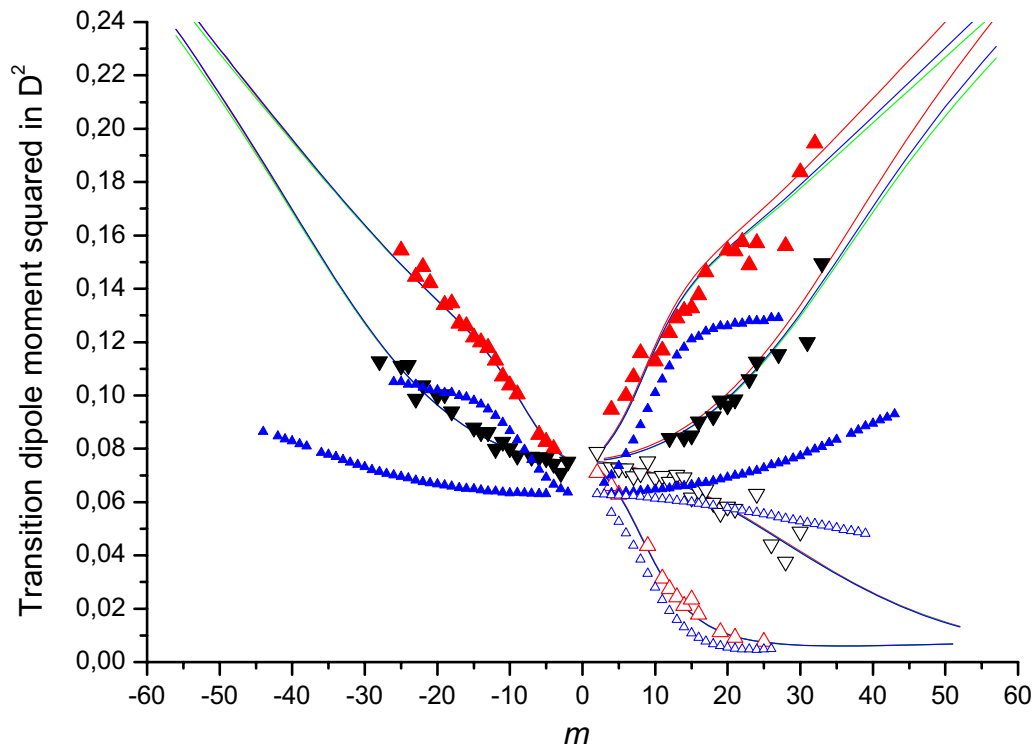
Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches of the  $00002\ 0 - 00001\ 0\ 1$  band ( $2\nu_5^0 - \nu_5^1$ ) plotted versus  $m$ . Black line is from HITRAN [11]. Black and red triangles are measurements from Ref. [13] and the present work respectively (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].



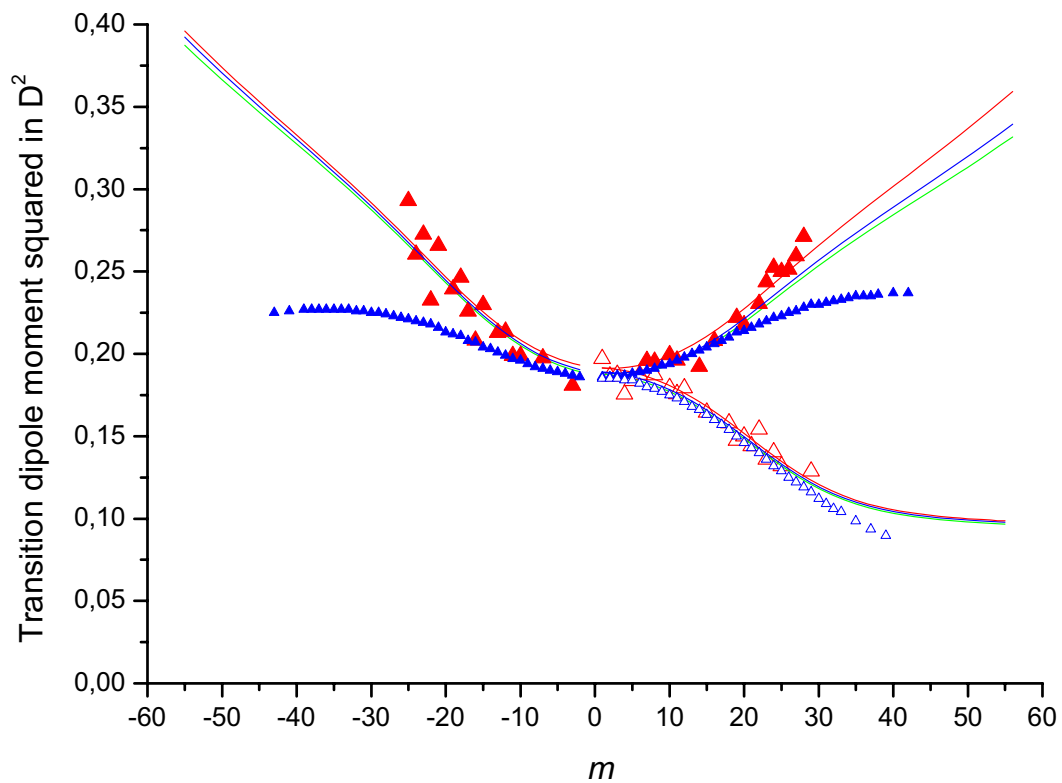
Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00002\ 0\ 2 - 00001\ 0\ 1$  band plotted versus  $m$  (noted  $2v_5^2 - v_5^1$  in Ref. [13] as well as in HITRAN and GEISA databases). Black line is from HITRAN [11]. Black and red triangles are measurements from Ref. [13] and the present work respectively (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Upside triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas downside triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].



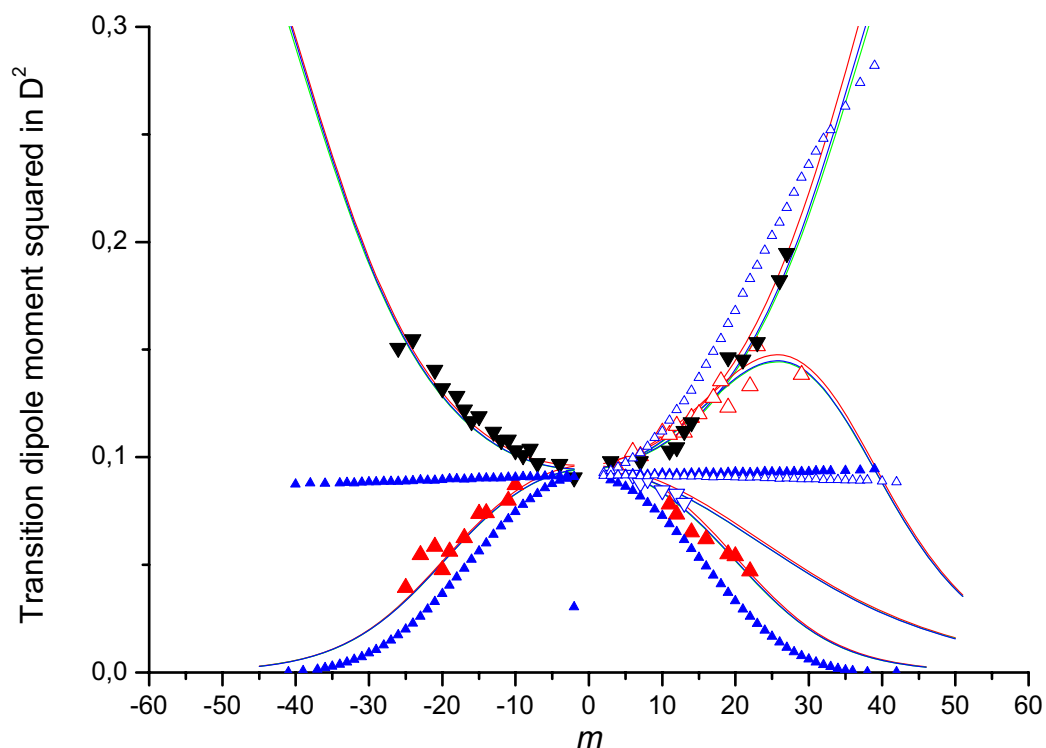
Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00011\ 1-1 - 00010\ 1\ 0$  band plotted versus  $m$  (noted  $(v_4+v_5)_+ - v_4^1$  and  $(v_4+v_5)_- - v_4^1$  in Ref. [13] as well as in HITRAN and GEISA databases). Black line is from HITRAN [11]. Black and red triangles are measurements from Ref. [Jac2001] and the present work respectively (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Upside triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas downside triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].



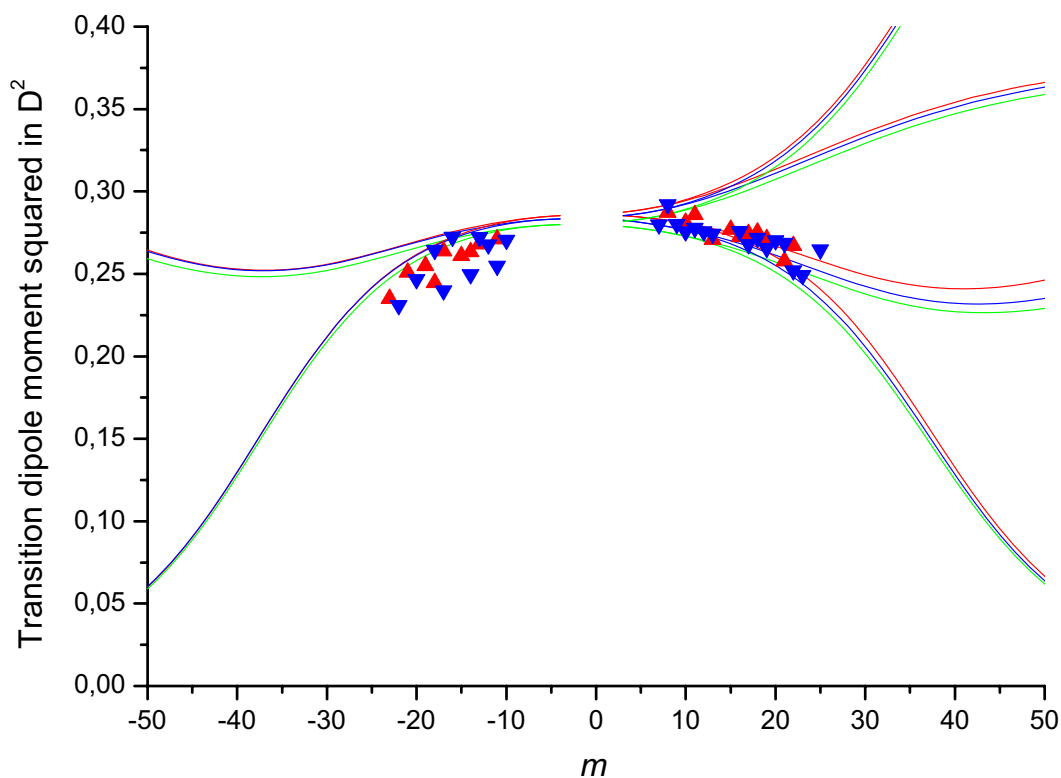
Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the 00021 2-1 – 00020 2 0 band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas black triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].



Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches of the 00003 0 1 – 00002 0 0 band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

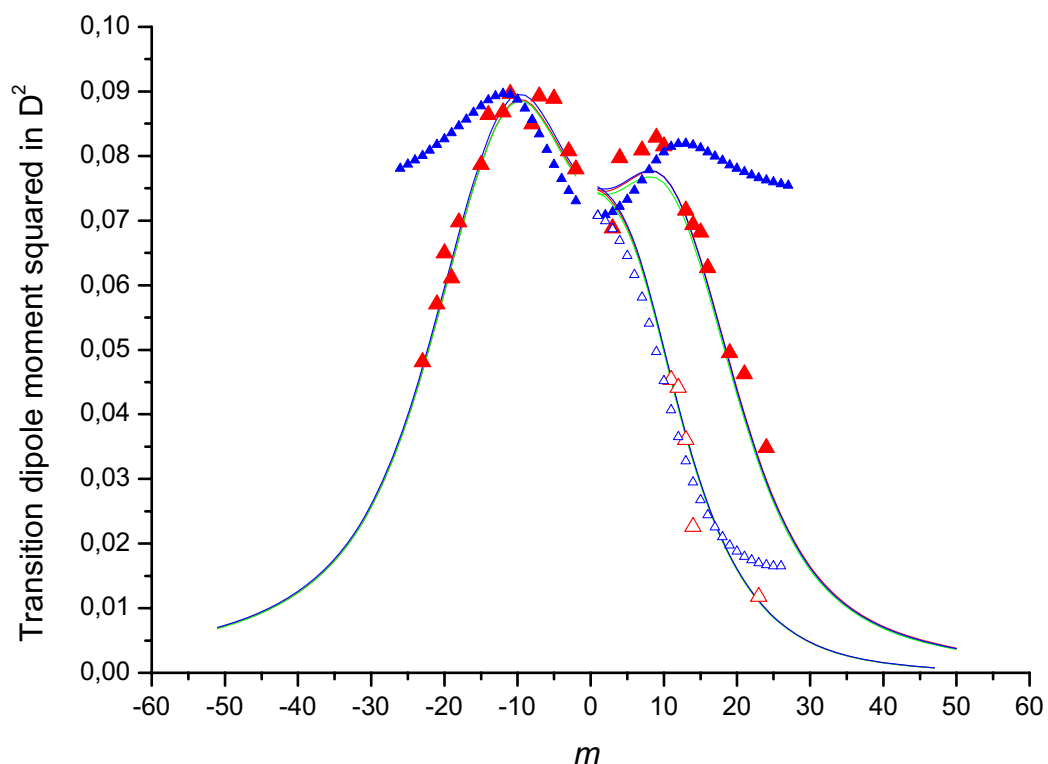


Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00003\ 0\ 1 - 00002\ 0\ 2$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ -and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas black triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

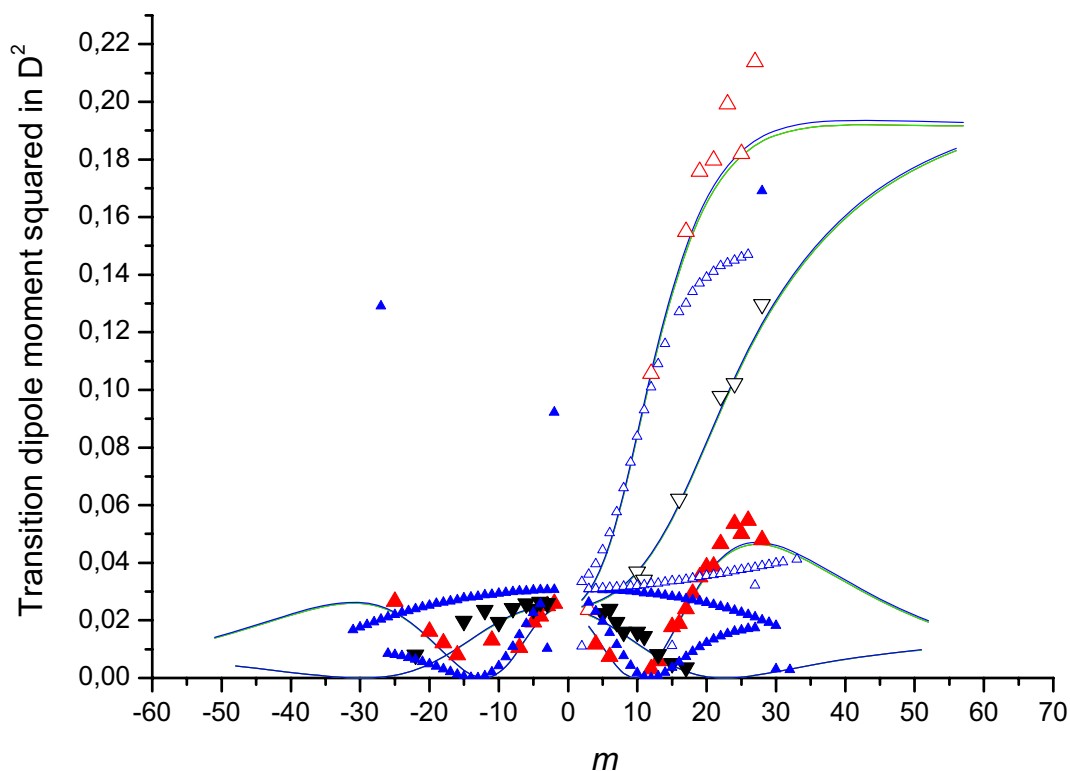


Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00003\ 0\ 3 - 00002\ 0\ 2$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$  and  $R_{ee}$ -branches whereas blue triangles are for  $P_{ff}$  and  $R_{ff}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Not Found in EXOMOL.

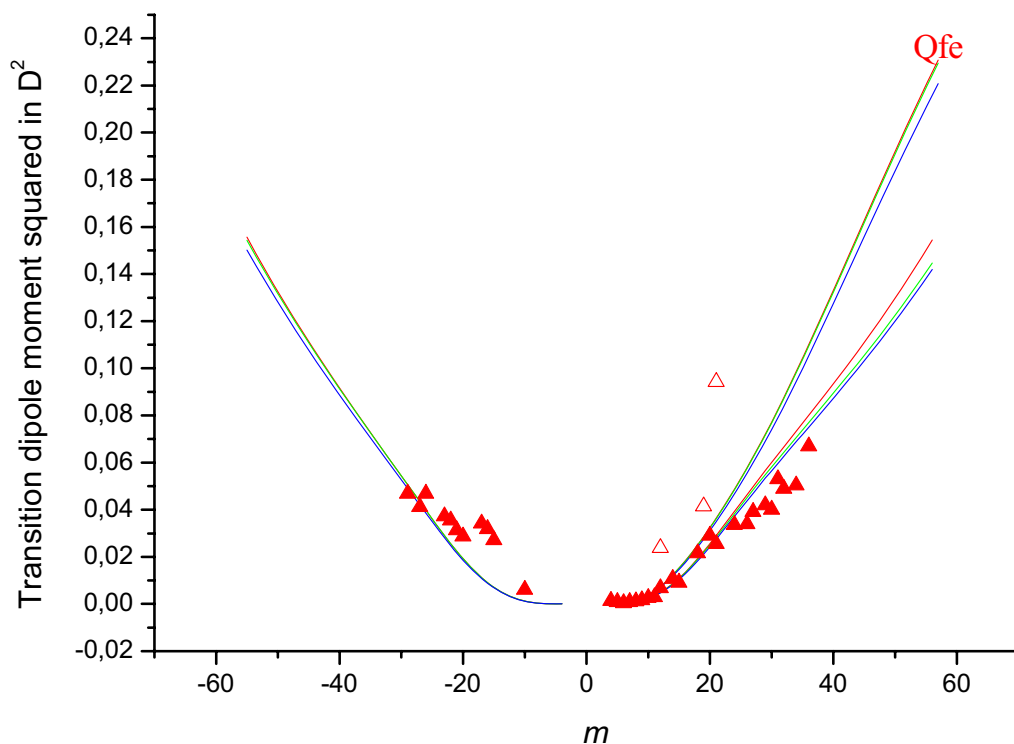




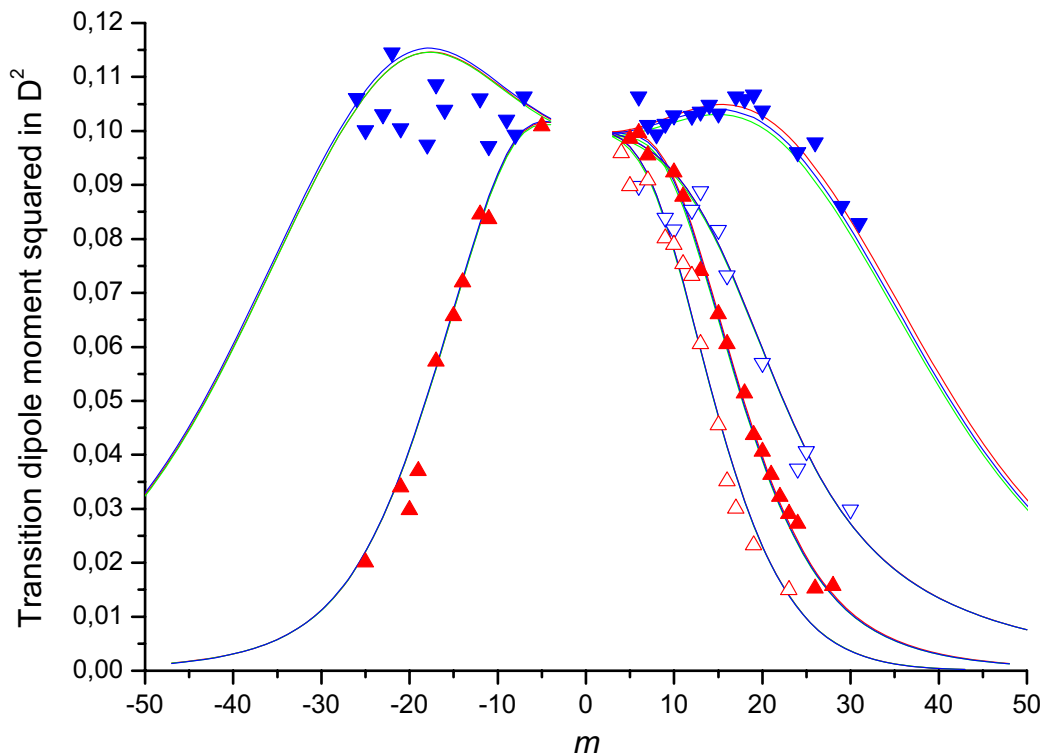
Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches of the **00021 0 1 – 00020 0 0** band plotted versus  $m$ . Red triangles are measurements from this work for the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches (open triangles for  $Q$ -branch and solid ones for  $P$ - and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].



Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00021\ 0\ 1 - 00020\ 2\ 0$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas black triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

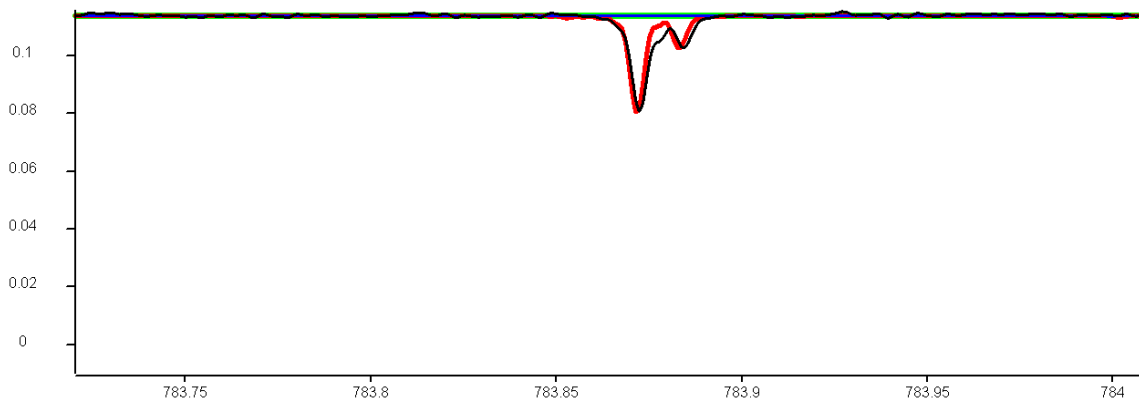


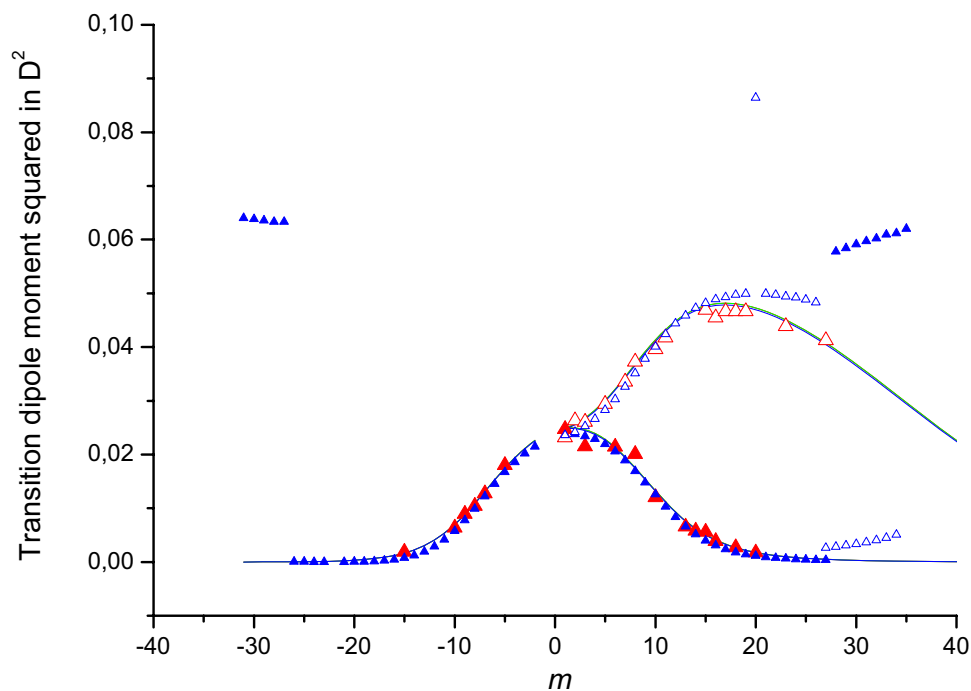
Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches of the **00021 2 1 – 00020 0 0** band plotted versus  $m$ . Red triangles are measurements from this work for the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches (open triangles for  $Q$ -branch and solid ones for  $P$ - and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Not Found in EXOMOL.



Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00021\ 2\ 1 - 00020\ 2\ 0$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas blue triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Not Found in EXOMOL.

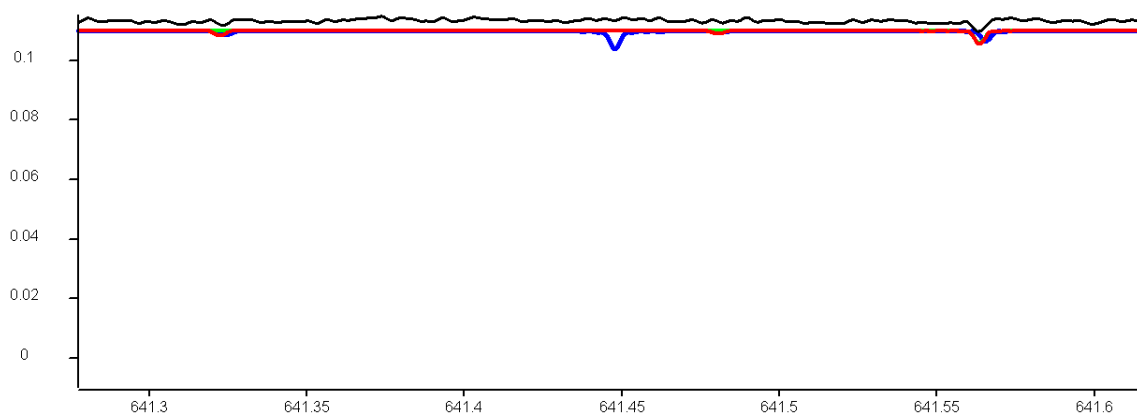
I checked by plotting synthetic spectra around the R20f quite visible in our exp spectra at  $783.8717\ \text{cm}^{-1}$ , the most intense below (in blue EXOMOL, green HITRAN 2016, red ASD).

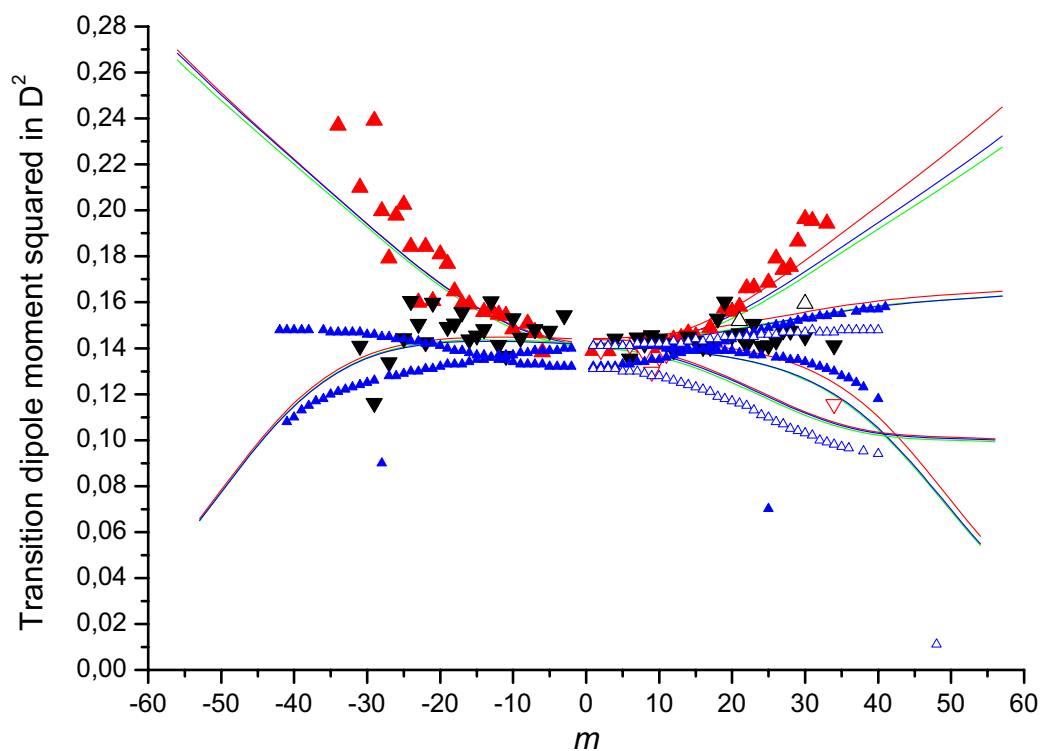




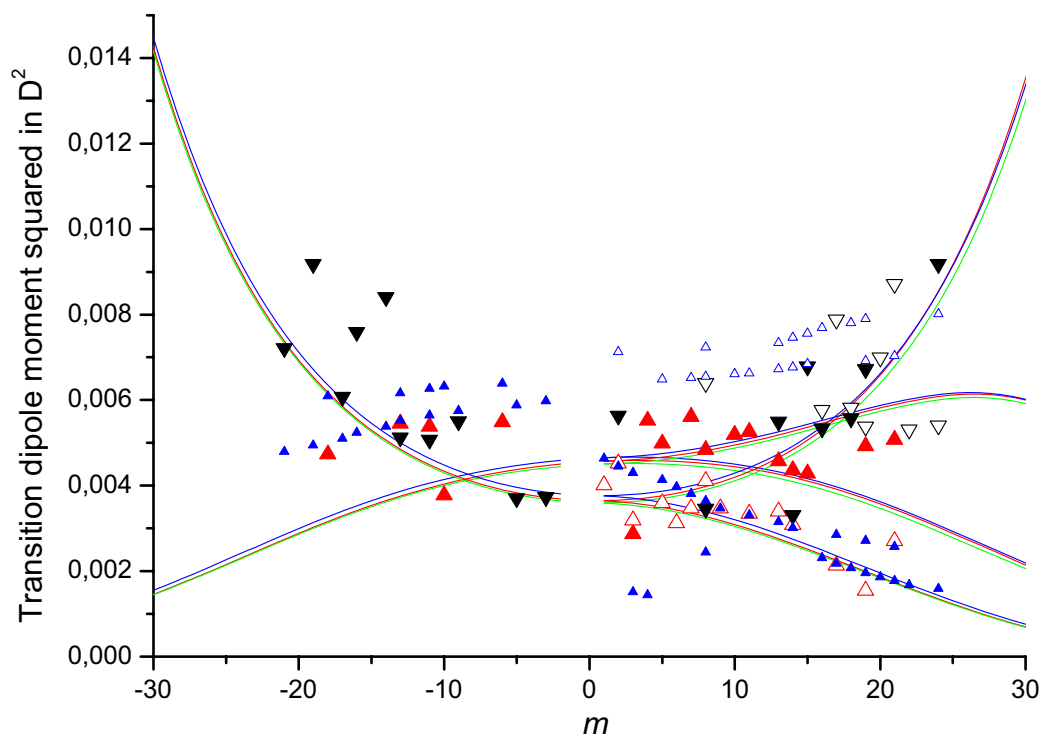
Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{je}$ -branches of the **00021 2-1 – 00020 0 0** band plotted versus  $m$ . Red triangles are measurements from this work for the  $P_{ee}$ ,  $R_{ee}$  and  $Q_{je}$ -branches (open triangles for  $Q$ -branch and solid ones for  $P$ - and  $R$ -branches). Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

Line P27e at 641.4486 has strange R2 value around 0.06 D2. And indeed the EXOMOL synthetic spectrum (in blue) presents a line that is not observed.

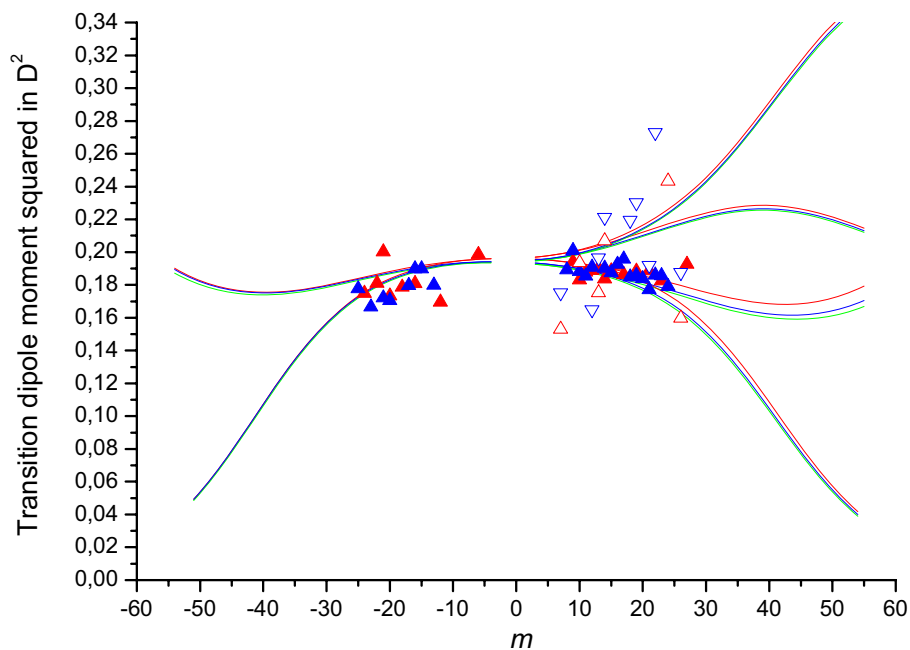




Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00012\ 1\ 0 - 00011\ 1-1$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas black triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

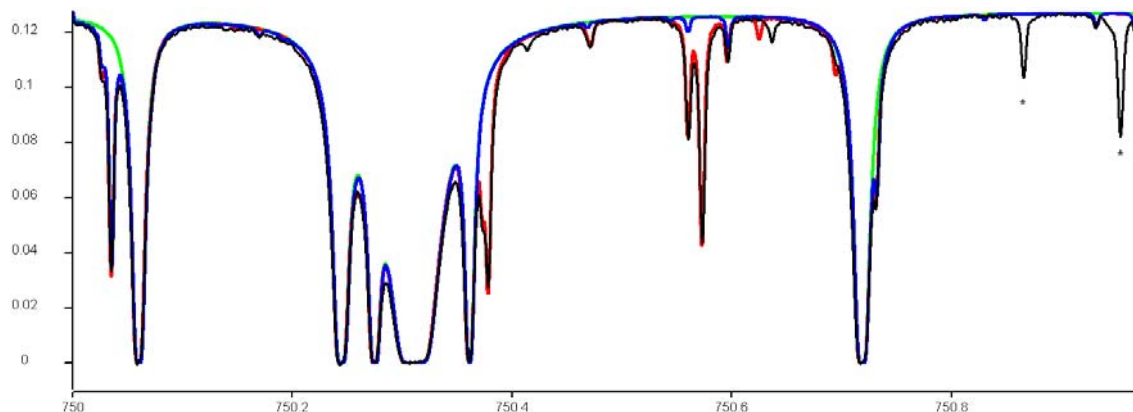


Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00012-1\ 2 - 00011\ 1-1$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas black triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].

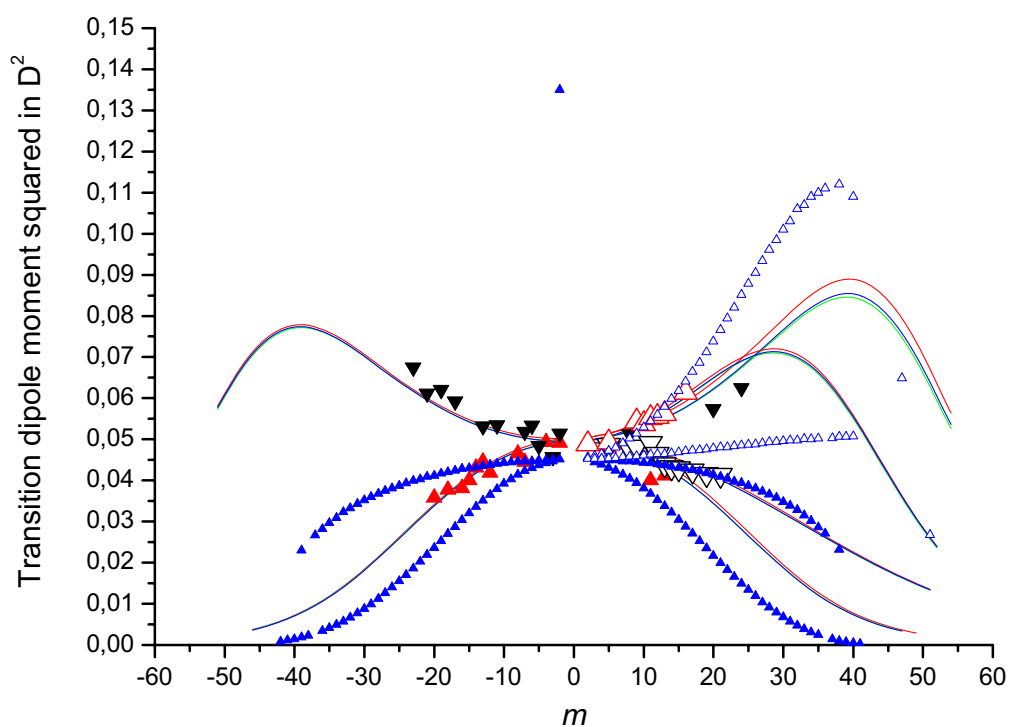


Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00012\ 1\ 2 - 00011\ 1\ 1$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas blue triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Not Found in EXOMOL.

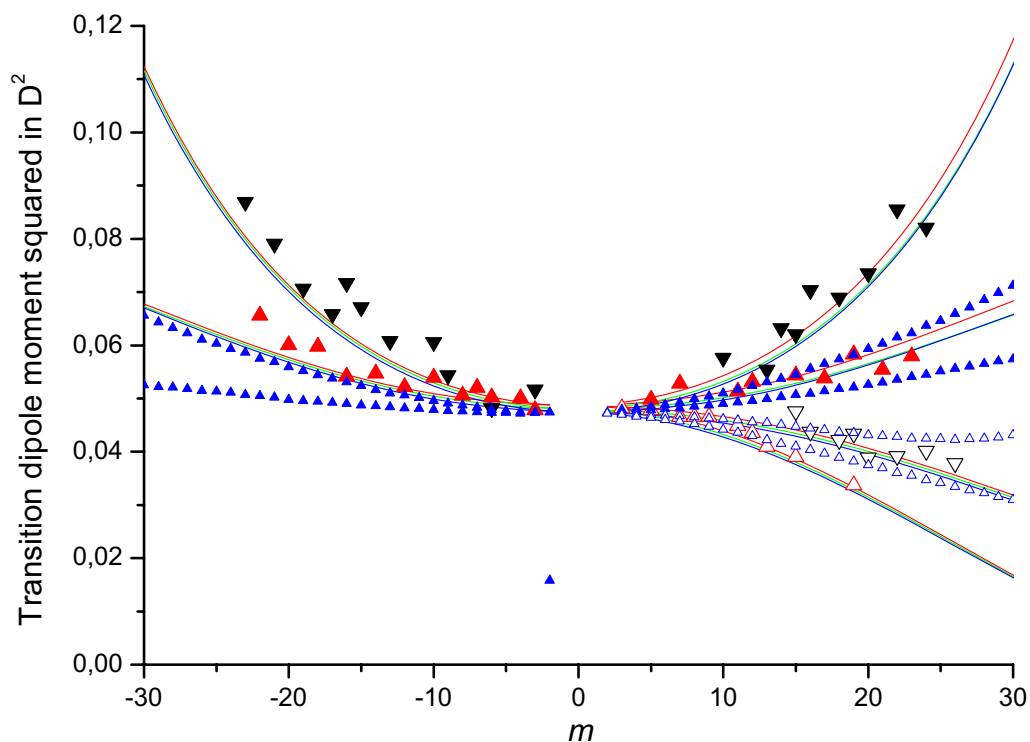
Below at  $750.378\ \text{cm}^{-1}$ , the Rff7 of this band observed in exp spectrum (black line) is reproduced by our line list (ASD in red), but not by HITRAN or EXOMOL. Around  $750.56$ , two lines are also missing in EXOMOL, the Ree8 and Rff8 of  $000\ 0\ 3\ 0\ 3\ \_u\ 000\ 0\ 2\ 0\ 2\ g$  band also missing in EXOMOL. The transitions with stars correspond to  $^{12}\text{C}^{13}\text{H}_2$  transitions.







Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00012\ 1\ 0 - 00011\ 1\ 1$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ - and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas blue triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].



Transition dipole moment squared of transitions belonging to the  $P_{ee}$ ,  $R_{ee}$ ,  $P_{ff}$ ,  $R_{ff}$ ,  $Q_{ef}$  and  $Q_{fe}$ -branches of the  $00012-1\ 2 - 00011\ 1\ 1$  band plotted versus  $m$ . Triangles are measurements from the present work (open for  $Q$ -branch and solid for  $P$ -and  $R$ -branches). Red triangles are for  $P_{ee}$ ,  $R_{ee}$  and  $Q_{fe}$ -branches whereas black triangles are for  $P_{ff}$ ,  $R_{ff}$  and  $Q_{ef}$ -branches. Calculations obtained using parameters of effective dipole moment are in blue line when using parameters from Ref. [4], in green line when using parameters from Ref. [1], and in red line when using parameters from this work (see Table 4). Plotted in blue symbols (open triangles for  $Q$  branch and solid triangles for  $P$  and  $R$  branches) are transition dipole moment squared from EXOMOL [26].