

Accurate laboratory rest frequencies of vibrationally excited CO up to $v = 3$ and up to 2 THz

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Received 17 December 2008 / Accepted 6 February 2009

ABSTRACT

Aims. Astronomical observations of (sub)millimeter wavelength pure rotational emission lines of the second most abundant molecule in the Universe, CO, hold the promise of probing regions of high temperature and density in the innermost parts of circumstellar envelopes.

Methods. The rotational spectrum of vibrationally excited CO up to $v = 3$ has been measured in the laboratory between 220 and 1940 GHz with relative accuracies up to 5.2×10^{-9} , corresponding to ~ 5 kHz near 1 THz.

Results. The rotational constant B and the quartic distortion parameter D have been determined with high accuracy and even the sextic distortion term H was determined quite well for $v = 1$ while reasonable estimates of H were obtained for $v = 2$ and 3.

Conclusions. The present data set allows for the prediction of accurate rest frequencies of vibrationally excited CO well beyond 2 THz.

Key words. molecular data – methods: laboratory – techniques: spectroscopic – radio lines: ISM – radio lines: stars – ISM: molecules

1. Introduction

Carbon monoxide, CO, is the second most abundant molecule in the universe after hydrogen, H₂. Observations of CO are commonly employed to obtain information on the H₂ abundance since H₂ is usually difficult to detect directly because it does not have any permanent or transition dipole moment, and its quadrupole transitions are very weak and at high energies. Usually, transitions of the main isotopologue ¹²C¹⁶O (in the following, unlabeled atoms designate ¹²C or ¹⁶O) are recorded for this purpose, but frequently also transitions of ¹³CO or even C¹⁸O are studied because transitions of these isotopic species are less affected by opacity. Even less abundant isotopologues have been detected in space, including recently the least abundant ¹³C¹⁷O (Bensch et al. 2001).

Very accurate ground state rest frequencies have been obtained in the laboratory for CO (Winnewisser et al. 1997) as well as for its isotopologues (Zink et al. 1990; Klapper et al. 2000a,b, 2001, 2003; Cazzoli et al. 2002, 2003, 2004; Puzzarini et al. 2003). The lower- J transitions were usually measured in sub-Doppler resolution such that the relative accuracies of the measurements reach $\sim 10^{-9}$.

Interestingly, the only experimental article to report measurements of transitions of vibrationally excited CO seems to be a paper by Bogey et al. (1986) on CO $J = 2-1$ transitions for several vibrational states between $v = 5$ and $v = 40$. One reason for this lack of measurements is probably the high vibrational energy of 2143 cm⁻¹ or 3086 K for $v = 1$ (Coxon & Hajigeorgiou 2004) which makes excited states of CO difficult to populate. Another reason is likely the small dipole moment

of 0.10980 (3) D (Muenther 1975) which even decreases for the first few vibrational states by about 0.025 D for each additional vibrational excitation until the dipole moment switches polarity between $v = 4$ and 5 (Goorvitch 1994), see also Fig. 1. Coxon & Hajigeorgiou (2004) reviewed rather extensively not only rotational data but also a plethora of infrared studies. Their work permits very good predictions of the rotational transitions of CO in excited vibrational states.

Apart from molecular clouds, CO is also an important constituent of the circumstellar envelopes of mass-losing stars on the asymptotic giant branch (AGB) of both carbon-rich ([C/O] abundance ratio >1) and oxygen-rich ([C/O] < 1) objects. Moreover, unlike other species, it does not condense into dust grains and exists throughout the envelope until it is destroyed by ultraviolet radiation in its tenuous outer portions (see, e.g., Lafont et al. 1982; Cherchneff & Barker 1992). Thus, emission from its strong rotational lines dominate the cooling and, thus, the thermal balance of an envelope (see, e.g. Crosas & Menten 1997).

CO is produced abundantly under the thermodynamical equilibrium conditions in the atmospheres of such AGB stars, even at photospheric temperatures, ~ 2000 K, (Tsuiji 1964) and is consequently observed easily via its near-infrared vibrational-rotational transitions. Toward the archetypical, high mass-loss C-rich object IRC+10216, Sahai & Wannier (1985) observed vibration-rotational emission and absorption in the 4.6 μ m fundamental band, while Keady et al. (1988) additionally observed absorption from the first overtone band at 2.3 μ m.

Given the extremely high temperature and density in the region from which the above transitions arise, one might also expect to find pure rotational emission to be observable from it.

However, pure rotational transitions of CO in excited vibrational states had not been reported in the astronomical literature until very recently (Patel et al. 2009a, see also below), while rotational lines of molecules chemically related to CO, although with considerably lower vibrational energies, E_{vib} , have been found toward IRC+10216. These include CS (Turner 1987a) and SiS (Turner 1987b), which have $E_{\text{vib}} = 1272 \text{ cm}^{-1}$ (Müller et al. 2005) and 744 cm^{-1} (Müller et al. 2007), respectively; see also (Patel et al. 2009b). Moreover, vibrationally excited SiO maser emission has been detected from the envelopes of many hundreds of oxygen-rich long period variable stars and red supergiants, and both its $v = 1$ and 2 ($E_{\text{vib}} = 1230$ and 2447 cm^{-1} , respectively (Sanz et al. 2003)) $J = 1-0$ lines are observed even toward three high-mass protostellar objects embedded in molecular clouds (Zapata et al. 2009).

It is noteworthy that for all three of these molecules transitions of even higher excited vibrational states and rare isotopologues have been observed, namely $v = 4$ for SiO (Cernicharo et al. 1993) and SiS (Agúndez et al. 2008; Patel et al. 2009b), and $v = 3$ for CS (Agúndez et al. 2007) and ^{29}SiO (Gonzalez-Alfonso et al. 1996).

In the case of CO, Scoville & Solomon (1978) reported the detection of its $v = 1$, $J = 1-0$ transition toward IRC+10216. However, that “detection” was soon rebutted by Cummins et al. (1980). They identified the observed spectral feature as due to C_4H , whose frequency is very close indeed to that of the $v = 1$, $J = 1-0$ CO line. Moreover, C_4H exhibits generally strong lines in this source. It took roughly 30 years until a genuine detection of pure rotational CO emission lines ($v = 1$, $J = 2-1$ and $3-2$) with the Submillimeter Array was reported (of course in IRC+10216) very recently by Patel et al. (2009a).

We have recorded rotational transitions of vibrationally excited CO belonging to four isotopic species, CO, ^{13}CO , C^{18}O , and $^{13}\text{C}^{18}\text{O}$ up to $v = 3$ in excitation and up to 2 THz in frequency. In order to facilitate the detection of radio lines of vibrationally excited CO, we present here our results for the main CO isotopologue. The full account of our work, which involves a combined fit with a variety of rotational and ro-vibrational data, will be presented elsewhere (Gendriesch et al., in preparation).

2. Experimental details

Transitions with frequencies below 1 THz have been recorded with the Cologne Terahertz Spectrometer (CTS) which has been described in detail by Winnewisser et al. (1994). It uses broadband tunable, phase-locked backward-wave oscillators (BWOs) as powerful sources, and a magnetically tuned, liquid helium cooled hot-electron InSb bolometer as detector. The Cologne Sideband Spectrometer for Terahertz Application (COSSTA) was employed for the measurements between 1.75 and 2.0 THz. In this case, the terahertz radiation was generated by mixing of a fixed-frequency far-infrared laser and a BWO as tunable sideband generator. An InSb bolometer was used again as detector. The spectrometer system has been described in greater detail in Gendriesch et al. (2000).

The pressure of CO in the 3 and 2 m long absorption cells was in the 1–4 Pa range. The measurements were carried out in a static mode or in a slow flow through the cell. One has to keep in mind that in the terahertz region a flow speed of several m/s causes frequency shifts of a few tens of kilohertz.

The smaller dipole moment in $v = 1$ compared with the vibrational ground state reduces the intensities by about 40%; in $v = 3$ the intensity is decreased by almost one order of magnitude, see Fig. 1. The high excitation energy reduces the

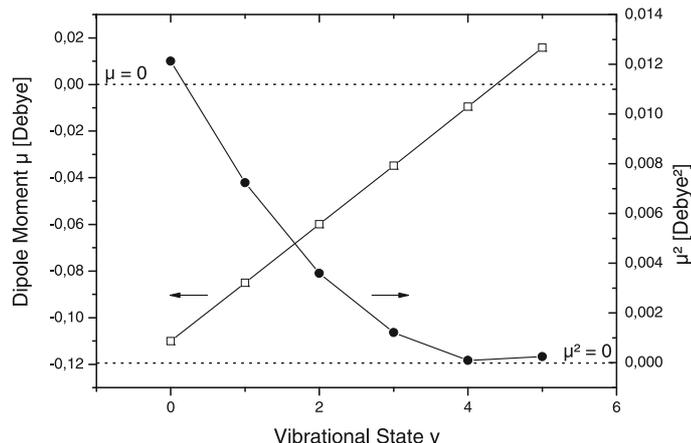


Fig. 1. The open squares with the y -scale to the left show the decreasing dipole moment from $v = 0$ to 4. Between $v = 4$ and 5 the dipole moment switches polarity from the negative polarity being at the C atom for $v \leq 4$ to the negative polarity being at the O atom for $v \geq 5$. The filled circles show the much larger effect in the squares of the dipole moments which are proportional to the intensities of the lines. The y -scale is on the right hand side.

intensities much more, by about 4.5 orders of magnitude for each vibrational quantum at 300 K. For these reasons, vibrationally excited CO could not be observed even under very favorable conditions and with long integration times at room temperature for $v > 1$. A glow discharge effectively increased the vibrational temperature such that lines of vibrationally excited CO could be recorded below 1 THz with the CTS. The available source power at 2 THz is considerably lower with COSSTA. Therefore, a modulation of the discharge (on/off) at ~ 5 Hz was employed in addition to the usual frequency modulation to reduce baseline effects and to improve the spectrometer stability. The measurements were carried out similarly to those on HNC (Thorwirth et al. 2000) and DNC (Brünken et al. 2006) in their ground and $v_2 = 1$ excited vibrational states.

The high accuracy of a transition frequency in our measurements depends very much on the flatness of the baseline, the absence of other, interfering lines in the vicinity, and on a high signal-to-noise ratio (S/N). Relative accuracies of about 10^{-8} have been reached for strong, isolated lines recorded in Doppler mode up to 1 THz as well as near 2 THz fairly commonly as demonstrated in the investigations of, e.g., H_2CO (Brünken et al. 2003) and SO_2 in its $v_2 = 0$ and 1 vibrational states (Müller & Brünken 2005). Under favorable conditions, better accuracies can be reached even in Doppler-limited measurements, as shown in the studies of HCN, $v_2 = 1$ (Thorwirth et al. 2003), and SiS (Müller et al. 2007). The study of rotational transitions of CO by Varberg & Evenson (1992) demonstrates that such high relative accuracies are possible also in other laboratories and at even higher frequencies (beyond 4 THz); see also Winnewisser et al. (1997) for an updated list of these frequencies and uncertainties. It is worthwhile mentioning that in sub-Doppler measurements relative accuracies of better than 10^{-9} have been reached e.g. in measurements of CO (Winnewisser et al. 1997) and ^{13}CO (Cazzoli et al. 2004).

3. Analysis and discussion

Predictions of the CO, $v = 1$ to 3 rotational spectrum were based initially on infrared measurements as summarized, e.g. in Coxon & Hajigeorgiou (2004). All but the very weak $J = 1-0$

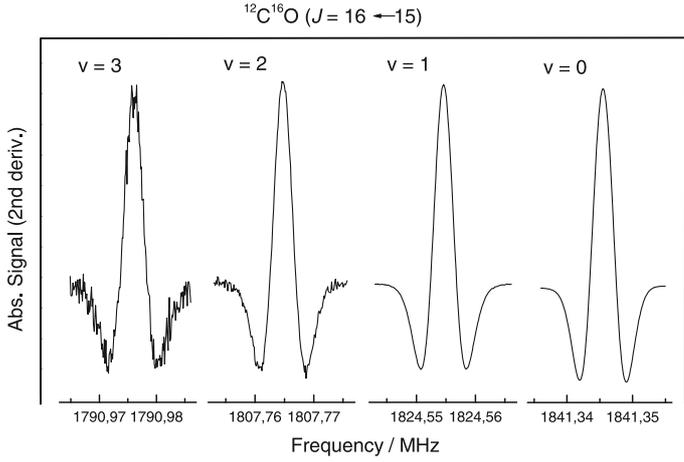


Fig. 2. The $J = 16 \leftarrow 15$ transitions are shown for $v = 0$ to 3. Note that the signal-to-noise ratio is very good even in the $v = 3$ state. The lower state energy of the transition is 6800 cm^{-1} , see Table 1. The lines appear as second derivatives of a Gaussian line-shape because of the $2f$ -modulation.

transition were recorded for $v = 1$ below 1 THz, fewer transitions were recorded for higher excited states. Two additional transitions could be recorded for each vibrational state with COSSTA near 2 THz. Figure 2 displays the very good signal-to-noise ratios obtained for the $v = 0$ to 3 states. Altogether between 9 and 4 experimental transition frequencies were obtained in the course of the present investigations. They are given in Table 1 together with their uncertainties. Also included are the calculated frequencies up to $J = 20\text{--}19$ with uncertainties, the lower state energies, and the line strengths $S\mu^2$.

The spectroscopic parameters have been determined in a weighted least-squares fit, and are listed in Table 2 together with those of the ground vibrational state from Winnewisser et al. (1997). The parameters B_v and D_v have been determined very well. Even the sextic centrifugal distortion term H_v has a standard deviation that is less than 10% of its value for $v = 1$, but it is barely determined for $v = 2$ and 3. The rotational constant B_v decreases with vibrational excitation, as is generally the case. The change in D_v is very small, but still determined with significance; the latter is not the case for H_v . The rms error of the fits, also known as the weighted standard deviation of the fit, are 0.561 for $v = 1$ and 2 suggesting the error estimates of the rest frequencies to be conservative. The rms error of 0.2 for $v = 3$ is not meaningful because 3 parameters have been determined from 4 transition frequencies only.

Table 1 provides calculated rest frequencies up to 2.3 THz. The small values of the predicted uncertainties suggest that reliable predictions of the rotational spectrum of CO $v = 1$ and probably 2 can be made up to at least 3 THz. Predictions to even higher frequencies will be available in the catalog section of the Cologne Database for Molecular spectroscopy¹ (Müller et al. 2001, 2005).

4. Astronomical outlook

As mentioned above, pure rotational emission from the $v = 1$, $J = 2\text{--}1$ and $3\text{--}2$ lines has been detected towards the hot, innermost circumstellar envelope of the high mass-loss carbon AGB star IRC+10216 (Patel et al. 2009a). Potentially, other vibrationally excited rotational lines from CO are detectable toward

Table 1. Measured transition frequencies^a (MHz) of vibrationally excited CO, transition frequencies^a (MHz) calculated from the final set of spectroscopic parameters, lower state energies E_{l_0} (cm^{-1}) and line strengths $S\mu^2$ (10^{-3} D^2).

J''	$v = 1$			
	measured	calculated	E_{l_0}	$S\mu^2$
0	–	114 221.7523 (13)	2143.271	7.228
1	228 439.074 (25)	228 439.1008 (24)	2147.081	14.444
2	342 647.636 (20)	342 647.6421 (33)	2154.701	21.638
3	456 842.977 (10)	456 842.9726 (37)	2166.131	28.795
4	571 020.677 (12)	571 020.6895 (37)	2181.369	35.909
5	685 176.392 (5)	685 176.3902 (34)	2200.416	42.960
6	799 305.677 (10)	799 305.6731 (32)	2223.271	49.950
7	913 404.136 (5)	913 404.1370 (40)	2249.933	56.851
8	–	1 027 467.3817 (59)	2280.401	63.657
9	–	1 141 491.0083 (84)	2314.674	70.366
10	–	1 255 470.619 (11)	2352.750	76.954
11	–	1 369 401.816 (13)	2394.628	83.431
12	–	1 483 280.205 (15)	2440.306	89.760
13	–	1 597 101.393 (15)	2489.783	95.941
14	–	1 710 860.986 (13)	2543.057	101.97
15	1 824 554.595 (10)	1 824 554.5953 (83)	2600.125	107.85
16	1 938 177.832 (10)	1 938 177.8318 (92)	2660.986	113.51
17	–	2 051 726.309 (23)	2725.636	119.04
18	–	2 165 195.644 (46)	2794.074	124.33
19	–	2 278 581.453 (77)	2866.298	129.41
$v = 2$				
0	–	113 172.3761 (31)	4260.062	3.588
1	226 340.341 (20)	226 340.3489 (56)	4263.837	7.169
2	339 499.521 (20)	339 499.5151 (72)	4271.387	10.732
3	–	452 645.4717 (76)	4282.712	14.270
4	565 773.818 (10)	565 773.8161 (69)	4297.810	17.778
5	678 880.140 (10)	678 880.1462 (66)	4316.682	21.240
6	791 960.061 (25)	791 960.0603 (98)	4339.327	24.658
7	905 009.183 (25)	905 009.158 (16)	4365.744	28.017
8	–	1 018 023.038 (25)	4395.932	31.304
9	–	1 130 997.302 (34)	4429.890	34.525
10	–	1 243 927.553 (42)	4467.616	37.665
11	–	1 356 809.392 (48)	4509.109	40.719
12	–	1 469 638.425 (50)	4554.367	43.677
13	–	1 582 410.258 (45)	4603.389	46.526
14	–	1 695 120.497 (32)	4656.172	49.273
15	1 807 764.748 (15)	1 807 764.752 (18)	4712.716	51.907
16	1 920 338.703 (80)	1 920 338.633 (48)	4773.016	54.408
17	–	2 032 837.75 (11)	4837.072	56.797
18	–	2 145 257.73 (21)	4904.880	59.032
19	–	2 257 594.17 (33)	4976.438	61.146
$v = 3$				
0	–	112 123.087 (12)	6350.439	1.207
1	–	224 241.770 (22)	6354.179	2.410
2	–	336 351.644 (30)	6361.659	3.603
3	–	448 448.307 (35)	6372.879	4.781
4	–	560 527.354 (37)	6387.837	5.939
5	672 584.390 (40)	672 584.384 (37)	6406.534	7.075
6	–	784 614.995 (40)	6428.969	8.182
7	896 614.750 (100)	896 614.785 (49)	6455.141	9.255
8	–	1 008 579.357 (65)	6485.049	10.290
9	–	1 120 504.311 (85)	6518.692	11.284
10	–	1 232 385.25 (11)	6556.068	12.232
11	–	1 344 217.78 (12)	6597.176	13.132
12	–	1 455 997.51 (13)	6642.014	13.976
13	–	1 567 720.05 (12)	6690.581	14.762
14	–	1 679 381.012 (94)	6742.874	15.488
15	1 790 976.008 (50)	1 790 976.004 (49)	6798.892	16.151
16	1 902 500.636 (100)	1 902 500.647 (96)	6858.633	16.747
17	–	2 013 950.56 (25)	6922.094	17.275
18	–	2 125 321.36 (47)	6989.272	17.728
19	–	2 236 608.68 (78)	7060.165	18.110

^a Numbers in parentheses are one standard deviation in units of the least significant figures.

¹ Website: <http://www.astro.uni-koeln.de/cdms/>

Table 2. Spectroscopic parameters^a (MHz) of CO in excited vibrational states compared with that in the ground vibrational state.

Parameter	$\nu = 0^b$	$\nu = 1$	$\nu = 2$	$\nu = 3$
B_0	57 635.968 019 (28)	57 111.243 12 (66)	56 586.555 00 (157)	56 061.910 5 (59)
$D_0 \times 10^3$	183.504 89 (16)	183.490 3 (71)	183.474 0 (234)	183.511 (63)
$H_0 \times 10^9$	171.68 (10)	177.9 (145)	163. (53)	241. (134)

^a Numbers in parentheses are one standard deviation in units of the least significant figures.

^b Winnewisser et al. (1997).

this and similar objects as well. The observations just reported, made with the Submillimeter Array, have an angular resolution of a few arc seconds and by no means resolve the emitting region, which is estimated to have more than 10 times smaller dimensions, $\sim 0''.2$ or ~ 30 AU for a distance of 150 pc (Crosas & Menten 1997). Resolving such small regions is outside the capabilities of existing (sub)millimeter interferometers.

However, the Atacama Large Millimeter Array (ALMA), currently under construction on the ~ 5000 m high Llano de Chajnantor in Chile, with its fifty 12 m diameter antennas will have a *much* higher angular resolution and a *much* larger collecting area than all existing instruments. Consequently, ALMA will have a vastly larger brightness temperature sensitivity, $\Delta T_B \propto [N(N-1)A_{\text{eff}}]^{-1}(\nu\theta_B)^{-2}(\Delta\nu)^{-1/2}$. Here N is the number of antennas (currently 50 planned and 64 as a goal), A_{eff} the effective area of a single antenna, ν the observing frequency, θ_B the synthesized beam size and $\Delta\nu$ the frequency resolution.

Using the online ALMA Sensitivity Calculator Tool² on ESO's ALMA WWW site we employ inputs equivalent to $\nu = 340$ GHz (comparable to the frequencies of vibrationally excited $J = 3-2$ CO line) and $\Delta\nu = 1$ MHz (corresponding to 0.88 km s^{-1} velocity resolution; i.e., appropriate for the $\sim 7 \text{ km s}^{-1}$ width of the lines observed toward IRC+10216), we calculate for 1.2 mm precipitable water vapor (mediocre weather conditions for the ALMA site) an rms flux density sensitivity of 3.2 mJy for 1 h of integration time. In an extended configuration, with a maximum baseline length, B_{max} , of 10 km, a beam of $FWHM \theta_B = c/(\nu B_{\text{max}})$ radians, or 22 milli arcseconds could be synthesized with ALMA (at $\nu = 340$ GHz; c is the speed of light). With this θ_B and the flux density sensitivity quoted above, we calculate that an rms brightness temperature sensitivity of 70 K could be achieved with ALMA at that resolution, which corresponds to a quarter of the diameter of IRC+10216's radio photosphere (Menten et al., in prep.).

Given that the brightness temperature of the gas emitting vibrationally excited CO is in high likelihood hotter than 1000 K (Patel et al. 2009a), spatially resolved imaging will allow studies of the distribution and dynamics of the hot material close to the stellar atmosphere of IRC+10216 and similar evolved stars. This will provide important insight into the origins of the mass loss process in such objects. These high resolution observations that will also easily image the radio photospheres of such near-by red giant stars (see, e.g., Reid & Menten 2007) will be eminently feasible since their stellar continuum emission will provide ample flux for self calibration.

Acknowledgements. This work has been supported by the Deutsche Forschungsgemeinschaft (DFG) via the collaborative research grant SFB 494. H.S.P.M. is grateful for recent support by the Bundesministerium für Bildung und Forschung (BMBF) administered through Deutsches Zentrum für Luft- und Raumfahrt (DLR). His support was aimed in particular at maintaining the CDMS.

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² <http://www.eso.org/sci/facilities/alma/observing/tools/etc/>