1972ApJ...177..609T

THE ASTROPHYSICAL JOURNAL, 177:609–617, 1972 November 1
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DETECTION OF THE 4_1–3_0 (E_2) LINE OF INTERSTELLAR METHYL ALCOHOL

B. E. TURNER AND M. A. GORDON
National Radio Astronomy Observatory,* Green Bank, West Virginia

AND

G. T. WRIXON
Bell Telephone Laboratories, Incorporated, Crawford Hill Laboratory, Holmdel, New Jersey
Received 1972 March 20

ABSTRACT

The $4_1 \rightarrow 3_0$ (E_2) transition of methyl alcohol (CH_3OH) at 36,169 MHz has been detected with a total strength of $\gtrsim$120 flux units (f.u.) in two extended clouds in Sgr B2. The line is not present to a limit of $\sim$30 f.u. in Ori A or in several other sources. This new line is further evidence, in addition to that of the recently detected $5_1 \rightarrow 4_0$ (E_2) transition, that the interstellar CH_3OH molecule is anomalously excited in most, if not all, of the transitions that have so far been detected in the interstellar medium.

We give limits to other molecular lines searched for but not detected in the range 28–40 GHz.

I. INTRODUCTION

Methyl alcohol was discovered in the interstellar medium by means of emission from the *K*-doublet transition in the 1_1 (A) state at 834 MHz (Ball *et al.* 1970). The two sources were Sgr B2 and Sgr A. At 834 MHz, the beamwidth of the telescope was too large to resolve structural details of the continuum background or of the CH_3OH cloud; thus no details about the line excitation could be derived. Subsequently Barrett, Schwarz, and Waters (1971) observed at 25 GHz the series of five transitions $K = 2 \rightarrow 1$, $J = 4$ to 8 in the E_1 species. These lines appeared only in Ori A. Although there were small deviations in the relative intensities of these lines from those expected in LTE, Barrett *et al.* assumed the lines to form in LTE, which led to unexpectedly large densities of CH_3OH in Ori A ($\sim$0.25 cm^{-3}).

However, recent observations indicate that the excitation of CH_3OH is probably anomalous. Zuckerman *et al.* (1972) discovered the 5_1–4_0 (E_2) line at 84.5 GHz in Sgr B2 but not in Ori A. It was noted that the lifetimes (with respect to spontaneous emission) of the upper levels were larger than those of the lower levels for the 5_1–4_0 (E_2) transition as well as for the 25-GHz transitions observed by Barrett *et al.* Assuming the levels to be depopulated mainly by spontaneous emission, one might therefore expect a population inversion in these levels and, at least, weak maser action in the corresponding transitions. This hypothesis produces a distribution of intensities among the E_1 lines similar to that observed by Barrett *et al.*

This model predicts that the 4_1–3_0 (E_2) line at 36,169 MHz should be observable and, with considerably more doubt, the 7_2–8_1 (E_2) line at 37,704 MHz and the 4_0–3_1 (E_1) line at 28,316 MHz. This report presents the results of searches for all three of these transitions.

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II. OBSERVATIONS

The observations of the 4_1–3_0 (E_2) line were made with the 120-foot (36.6-m) Haystack antenna. Under ideal weather conditions, which applied to the present CH_3OH observations, the effective aperture efficiency at 35.0 GHz is $\eta_A = 12$ percent at elevation 45° and 10.8 percent at elevation 15°. The beam efficiency η_B at elevation 15° is estimated to be 14 percent, if we assume a Gaussian beam and standard illumination. These parameters include the effect of the radome, but are uncertain for two reasons. First, the efficiency depends strongly on the amount of moisture on the radome, the distribution of which may be asymmetrical and vary with position and time over the radome (W. Dent, private communication). Second, the sidelobes are as large as 10 dB and thus a Gaussian shape does not describe the beam well. Therefore, 14 percent is probably an upper limit to the beam efficiency. With $\eta_A = 10.8$ percent, the antenna sensitivity is $\sim$24 f.u. per °K antenna temperature for a point source. The full beam size is approximately 1′ at the 3-dB points. Extinction corrections of 25 percent have been applied to the data in table 2.

The receiver consisted of a balanced diode mixer with a matched intermediate-frequency amplifier. The system temperature ranged between 700° and 1300° K double-sideband, depending upon the frequency; its value at 36.2 GHz was $\sim$1000° K. The local oscillator frequency was phase-locked to a hydrogen maser and was accurate to within one part in $\sim 10^{11}$, or within 1 Hz for a local-oscillator range of 28 to 42 GHz. The spectrometer was the new 100-channel autocorrelation receiver of the Haystack Observatory; it was operated at a total bandwidth of 20 MHz, giving a resolution (with Hanning smoothing) of 500 kHz (4.1 km s^{-1} at 36,169 MHz). We took spectra of the total power on and off the source every 5 minutes; a Univac 490 computer differenced, normalized, and averaged these spectra on-line. No further manipulation of the data, such as removal of baseline effects, has been performed.

Figure 1 shows the spectra obtained at several positions on and about the OH position of Sgr B2 (R.A. = $17^h44^m11^s$; decl. = $-28°22'30''$ [1950]). The structure is evidently quite complicated; our sensitivity is adequate to establish two distinct velocity features, at 53.9 and 70.3 km s^{-1}, respectively. The line widths of these features are comparable, about 7 km s^{-1} at half-intensity. Both clouds are spatially extended relative to our 1′ beam, and occupy quite different positions. The 53.9 km s^{-1} cloud has a position $\sim$10″ east and 36″ south of the OH position, while the 70.3 km s^{-1} cloud is $\sim$15″ west and at least 70″ north of the OH position; observations at positions farther north are required to determine whether the 70.3 km s^{-1} cloud coincides with the continuum peak or with the peak in NH_3 (3,3) emission (Cheung *et al.* 1969).

At the same spatial resolution (1′), Penzias, Jefferts, and Wilson (1971) find CO velocity features at $\sim$56 and possibly 78 km s^{-1} as well as a strong feature at $\sim$93 km s^{-1}. The Sgr B2 region was not mapped sufficiently in the CO line to determine whether the first two CO features correspond in position to the two methyl alcohol clouds observed here at λ8.3 mm. At a resolution of $\sim$1.′5, Zuckerman *et al.* (1971) find that the 5_1–4_0 (E_2) line at 84.5 GHz comes from a source extended at least in declination by an amount that is consistent with the present observations. We assume the same source distribution at both frequencies in calculating the fluxes given in table 2. However, only one distinct velocity feature was observed at 84.5 GHz, centered at a velocity of 63 $\pm$ 3 km s^{-1} and having a full line width at half-intensity of 24 $\pm$ 3 km s^{-1}. These quantities are consistent with the velocity centroid (62.1 km s^{-1}) and overall line width (27.6 $\pm$ 3 km s^{-1} for both features together) of the present lines at λ8.3 mm. The failure to observe two clearly distinct velocity features at 84.5 GHz is probably due to a combination of a more limited spatial resolution and a smaller signal-to-noise ratio, rather than to any real differences in the emission brightness temperature as a function of velocity. Other single-dish observations of molecules in Sgr B2 provide no additional

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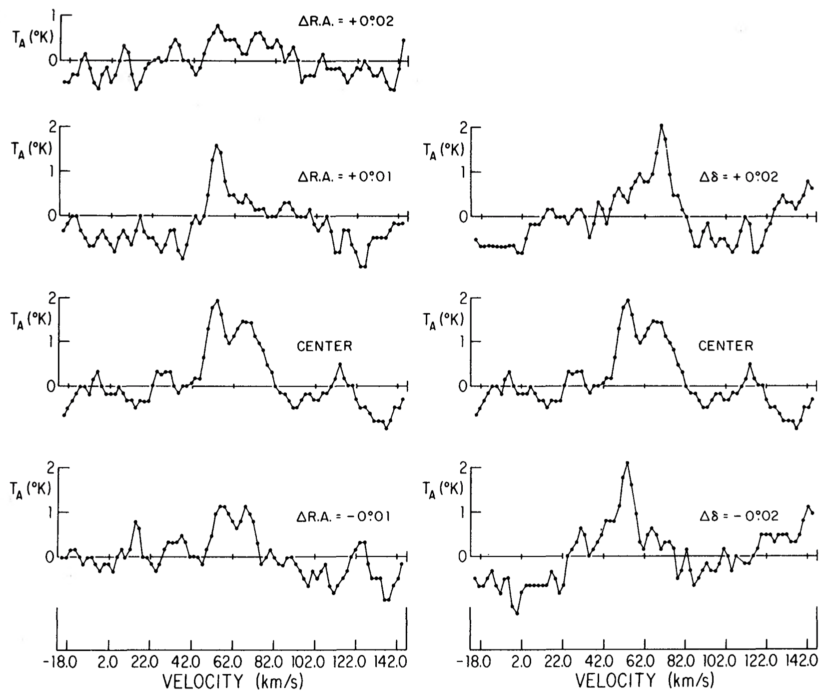

FIG. 1.—The $4_1 \rightarrow 3_0$ (E_2) line of methanol in Sgr B2. The assumed rest frequency is 36,169.24 MHz (Lees 1971), and the center position is that of Sgr B2(OH): R.A. $17^h44^m11^s$, decl. $-28°22'30''$ (1950.0).

information on the spatial fine structure, as they have all been made with larger beamwidths. The interferometric maps of H_2CO in Sgr B2 (Fomalont and Weliachew 1971) do not resemble the spatial distribution of the two CH_3OH clouds.

Table 1 summarizes the negative results obtained for the 4_1–3_0 (E_2) line in sources other than Sgr B2, and also for the 7_2–8_1 (E_2) and 4_0–3_1 (E_1) transitions of CH_3OH, as well as for several other molecules that were searched in the 28- to 40-GHz region.

III. DISCUSSION

Figure 2 shows the energy-level diagram for the A, E_1, and E_2 species of methyl alcohol, for values of K up to 3. The diagram indicates the transitions for which searches have been made. It should be noted that E_1 levels with $+K$ are degenerate with E_2 levels with $-K$. Therefore, there are two ways of labeling the torsion-rotation energy levels. We have adopted the convention that K is positive, in which case the levels are labeled as E_1 or E_2 according to the symmetry of the methyl group free-rotor basis functions under the C_3 group operations (Lees 1972). Alternatively, all levels can be called E_1 and labeled with positive and negative K values, because a $+K\ E_1$ level is degenerate with a $-K\ E_2$ level.

Lees (1972) has pointed out that $E_1 \leftrightarrow E_2$ transitions are forbidden under the symmetric-top-dipole selection rules $\Delta K = 0, \pm 1$. However, he remarks that molecular asymmetry can mix states with K's differing by 2, hence $\Delta K = \pm 3$ $E_1 \leftrightarrow E_2$ transitions are weakly allowed. For example, asymmetry mixing should weakly permit a $K = 2 \leftarrow -1$ E_1 transition, i.e., a transition $K = 2\ E_1 \leftarrow K = 1\ E_2$.

Table 2 summarizes the observational parameters of the transitions searched for in the two sources Ori A and Sgr B2. (The data have been assembled from Barrett *et al.* 1971, Zuckerman *et al.* 1971, and the present work.) There are obvious departures

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TABLE 1

NEGATIVE RESULTS

Molecule	Formula	Transition	Frequency (MHz)	Upper Limit (5 σ) to Antenna Temperature (°K)					Velocity Range† Searched (km s^{-1})
				Sgr B2	W51	Ori A	Globule*	IRC 10216	
Methanol	CH_3OH	$4_1 \rightarrow 3_0$ (E_2)	36,169.24‡	See text	0.60	(1.43)§	...	...	± 82.9
		$7_2 \rightarrow 8_1$ (E_2)	37,703.72‡	0.92	...	...	...	...	± 79.5
		$4_0 \rightarrow 3_1$ (E_1)	28,316.03‡	0.42	0.59	0.45	...	...	±105.9
Cyanamide	NH_2CN	2_{02}–1_{01}	39,989.42	...	...	0.64	...	...	± 75.0
Sulfur dioxide	SO_2	2_{02}–1_{01}	38,202.38	...	...	0.38	0.90	2.90	± 78.5
Methyl isocyanide	CH_3NC	2_{12}–1_{11}	40,210.48	0.94	0.44	0.38	...	...	± 74.6
		2_{02}–1_{01}	40,211.39	0.94	0.44	0.38	...	...	± 74.6
Methyl cyanide	CH_3CN	2_{02}–1_{01}	36,795.38	...	(0.90)	0.61	...	...	± 81.5
Ethyl cyanide	C_2H_5CN	4_{04}–3_{03}	35,722.18	(0.78)	...	(1.00)	(1.15)	...	± 83.9
Carbonyl sulfide	OCS	3_{03}–2_{02}	36,488.74	...	...	0.60	1.33	0.64	± 82.2
Acetaldehyde	CH_3CHO	2_{02}–1_{01}	38,512.40	...	...	0.38	1.08	...	± 77.8
Vinyl cyanide	CH_2CHCN	3_{03}–2_{02}	28,440.74	0.37	...	...	...	...	±105.4

* The observed position of this globule in NGC 2244 was $06^h28^m00^s$, $+04°58'18''$ as given by Sim (1968); this position has been found to be in error (Cordwell, private communication), and the observed position corresponds only with part of the H II region in NGC 2244.

† Centered on: 62 km s^{-1} for Sgr B2, 65.4 km s^{-1} for W51, 10.0 km s^{-1} for Ori A, 0.0 km s^{-1} for globule, −24.0 km s^{-1} for IRC 10216.

‡ Observed frequencies for CH_3OH kindly provided us by R. M. Lees (1971); they are close to the original values of Ivash and Dennison (1953).

§ Values in parentheses correspond to uncertain flux limits owing to cloudy weather.

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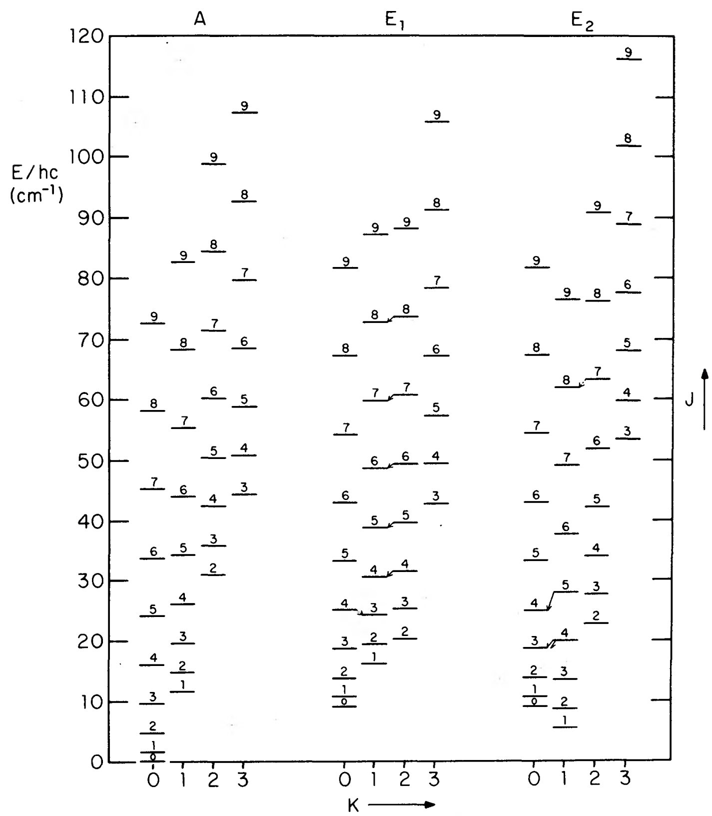

FIG. 2.—The energy-level scheme of methanol. Solid arrows denote lines detected in the interstellar medium; dotted arrows refer to those not present (see table 2 for upper limits). The $4_1 \rightarrow 3_0$ (E_2) line detected in this work is shown with a double arrow.

from the behavior expected for LTE excitation. First, E_1 transitions are anomalously strong relative to E_2 transitions in Ori A, and vice versa in Sgr B2. Second, the 4_0–3_1 (E_1) transition is anomalously weak compared with the other E_1 lines found in Ori A. In LTE the 4_2–4_1 and 4_0–3_1 lines should have comparable intensities: a ratio between 0.90 at $T = 30°$ K and 1.23 at $T = \infty$. Yet the observed ratio of brightness temperature is greater than 2. Third, in LTE the ratio of brightness temperatures of the 5_1–4_0 (E_2) and 4_1–3_0 (E_2) lines should be ≤ 2.8, independent of the temperature as long as it is not less than $\sim 30°$ K (the equality corresponds to optical thinness for both lines). The ratio should never be less than unity, which applies in the optically very thick case. However, the observed ratio of brightness temperatures has a value between 0.25 for the case where the source fills both beams, and 0.58 for the case where the source is pointlike. As mentioned above, the true distribution is closer to the filled-beam case.

Based principally upon the first point, Zuckerman *et al.* assumed that methyl alcohol was excited anomalously. They found that population inversions would be expected on the basis of the relative rates of population and depopulation of the various energy levels under the action only of spontaneous emission and of collisions, assuming that collision rates did not exceed spontaneous emission rates significantly. This model fits well the relative intensities of the detected E_1 lines in Ori A.

The data for the three additional transitions reported here provide another basis upon which to test the anomalous-excitation model. As in Zuckerman *et al.*, we have computed the sum of Einstein A-coefficients of all transitions that lead into and out of

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TABLE 2

OBSERVED STRENGTHS OF CH_3OH LINES (f.u.)

Transition	Ori A	Sgr B2
E_1 Lines		
$4_0 \rightarrow 3_1$.......	< 10*	< 20†
$4_2 \rightarrow 4_1$.......	20	...
$5_2 \rightarrow 5_1$.......	22	...
$6_2 \rightarrow 6_1$.......	34	...
$7_2 \rightarrow 7_1$.......	30	< 20†
$8_2 \rightarrow 8_1$.......	14	...
E_2 Lines		
$4_1 \rightarrow 3_0$.......	< 30*	130†
$5_1 \rightarrow 4_0$.......	<110*	180†
$7_2 \rightarrow 8_1$.......	...	< 50†

* Assumes a point source for the CH_3OH emission region.

† Calculated by convolving the two-cloud source distribution (assuming Gaussian distributions in intensity for each cloud) with the appropriate beam pattern. The fluxes refer to the sum of the contributions from the two clouds, and are rounded off to the nearest multiple of 10 f.u.

the levels involved in each of the transitions listed in Table 2.[1] These sums are given in table 3. The effects of internal rotation are rigorously taken into account in calculating the A-coefficients. (We have used computer-calculated values of the torsional contributions to the dipole moments, as kindly provided by R. M. Lees 1971.) Of course, the actual rates into and out of all of the relevant energy levels involve the product of level populations and A-coefficients, and hence cannot be calculated without detailed knowledge of the populations. A full solution of the statistical rate equations is therefore needed; as this requires such unknown quantities as collision cross-sections for neutral particles upon methyl alcohol, we have not considered such a detailed approach to be warranted. As a crude approach, one might assume that the populations will not vary as drastically from level to level as do the sum of the A-coefficients; then within this approximation, the entries in table 3 may be taken as being the rates. The expected population inversion, hence emission strength, is then proportional to the difference between $R_{in}(u) - R_{out}(u)$ and $R_{in}(l) - R_{out}(l)$, where $R_{in}(u)$ and $R_{out}(u)$ are respectively the sums of A coefficients in and out of the upper level and l refers to the lower level.

The values listed in table 3 explain qualitatively all of the observations of methyl alcohol in the E_1 and E_2 states. In the E_1 group, table 3 predicts that the 3_2–3_1 line will be found to be somewhat weaker than the other $K = 2 \rightarrow 1$ transitions near 25 GHz, but will probably be detectable. On the other hand, the 4_0–3_1 transitions ought to be much weaker even though it shares the same lower level and has an upper level (4_0) whose energy above ground (25.555 cm^{-1}) is virtually the same as that of the 3_2 level

[1] In computing the sums of Einstein A's, we have neglected contributions from weakly allowed transitions of the type $\Delta K = \pm 3$ (e.g., $K = 2\ E_1 \leftarrow K = 1\ E_2$) which arise from mixing of states by molecular asymmetry. The mixing is expected to be very slight, resulting in only very small electric-dipole moments for these transitions. The effect is analogous to the vibration-rotation interaction in NH_3, which induces dipole moments of order 10^{-5} debye for transitions described by $\Delta k = \pm 3$ (Oka *et al.*, 1971).

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TABLE 3

SPONTANEOUS RATES ASSOCIATED WITH OBSERVED TRANSITIONS OF CH_3OH*

	E_1 Lines			E_2 Lines	
Transition	$R_{in}(u) - R_{out}(u)$	$R_{in}(l) - R_{out}(l)$	Transition	$R_{in}(u) - R_{out}(u)$	$R_{in}(l) - R_{out}(l)$
$4_0 \rightarrow 3_1$...	138	−48	$4_1 \rightarrow 3_0$...	1826	−137
$3_2 \rightarrow 3_1$...	3006	−48	$5_1 \rightarrow 4_0$...	2133	−92
$4_2 \rightarrow 4_1$...	3349	−51	$7_2 \rightarrow 8_1$...	6892†	+3833
$5_2 \rightarrow 5_1$...	3674	−54			
$6_2 \rightarrow 6_1$...	4019	−59			
$7_2 \rightarrow 7_1$...	4403	−42			
$8_2 \rightarrow 8_1$...	4823	−27			

* Units are 10^{-6} per second.

† See text.

(25.1372 cm^{-1}). This conclusion is consistent with our failure to detect the 4_0–3_1 transition. In addition, the values given in table 3 for the $K = 2 \rightarrow 1$ transitions are consistent with the observed relative brightnesses of these lines in Ori A by Barrett *et al.* (1971).

For the E_2 lines, table 3 suggests that the 4_1–3_0 transition will have slightly weaker or comparable population inversion than the 5_1–4_0 transition, if the net populations are not falling off too rapidly with increasing energy for levels as high as the 5_1 level. Such a fall-off of population is, however, indicated by an observed brightness ratio $T_B(5_1–4_0)/T_B(4_1–3_0)$ that appears to be smaller than any value possible under LTE. Because of this, the prediction in table 3 for the 7_2–8_1 transition should not be taken literally; the large value of $R_{in}(u) - R_{out}(u)$ in this case stems solely from a very large A coefficient for the 7_3–7_2 transition; this contribution to $R_{in}(u)$ will, however, be realized only if there is significant population in the 7_3 level, and this is unlikely in view of its high energy above ground. If the contribution from the 7_3 level is indeed negligible, then $R_{in}(u) - R_{out}(u)$ for the 7_2–8_1 transition assumes the value 257, corresponding to a strongly anti-inverted population and an undetectable line. This latter situation seems to be indicated by the observations.

If the simple approach illustrated by table 3 is a reasonable approximation, then the observations indicate collisional rates in the methyl alcohol clouds that appear to populate significantly the levels up to ~ 70 cm^{-1} above ground in Ori A and ~ 28 cm^{-1} in Sgr B2. In the E_1 ladder seen in Ori A, observed decreases in line brightnesses from the predictions of table 3 are first apparent in the 8_2–8_1 line (the 8_2 level is 73.5 cm^{-1} above ground). In the E_2 ladder seen in Sgr B2, the observed decreases occur somewhere between the 4_1–3_0 and 5_1–4_0 transitions. Without more detailed calculations, and collisional cross-sections, one cannot estimate densities and temperatures separately.

Table 3 does not directly explain why E_1 transitions are anomalously strong relative to E_2 transitions in Ori A and vice versa in Sgr B2. A possible explanation lies in the fact that the observed E_2 lines lie lower in energy than the observed E_1 lines, as shown in figure 2. Assume that the density and kinetic temperature in the Sgr B2 cloud are significantly lower than in the Ori A source. Then the E_2 lines might not be observed in Ori A if the density were high enough to thermalize these transitions while the higher-lying E_1 transitions with $K = 2 \rightarrow 1$ are not thermalized.[2] Failure to observe the 4_0–3_1 (E_1) line in Ori A might be explained by the relatively unfavorable rates for

[2] This is possible because the collision cross-sections would not increase as fast with increasing J as the number of particles having the required energy would decrease, for temperatures of order 100° K or less.

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this transition given in table 3. Similarly, the E_1 lines with $K = 2 \rightarrow 1$ might be unobservable in Sgr B2 if the lower density and temperature there corresponded to too low a populating rate for these higher-lying transitions. In this regard we note that only the high-lying 7_2–7_1 (E_1) line has been searched in Sgr B2.

Two arguments suggest that such a simple picture cannot explain the observed differences in the Ori A and Sgr B2 sources. First, contrary to observation, one should expect to observe the 4_0–3_1 (E_1) line in Sgr B2, since it lies lower in energy than the 5_1–4_0 (E_2) transition which is observed in this source. Second, at least the 5_1–4_0 (E_2) transition ought to be observable in Ori A, given that the 4_2–4_1 (E_1) line is seen there, because the Einstein A coefficient is some 48 times larger for the 5_1–4_0 transition, and hence it is much less likely to be thermalized by collisions. A more stringent upper limit on the 5_1–4_0 (E_2) line may be important to decide this point.

It is tempting to conclude that the observations of methyl alcohol indicate the presence of processes that preferentially populate E_2 over E_1 levels in Sgr B2 and vice versa in Orion A. However, there are several problems with this interpretation. Different collisional species in the two sources might select the internal rotation states differently, but this seems inexplicable on the basis of current knowledge. Equally difficult to explain would be a selection of one species over the other during creation of the CH_3OH molecules on grain surfaces; no analogy seems possible with a mechanism that has been suggested to distinguish between the ortho and para forms of H_2 when it is formed on surfaces (Hollenbach and Salpeter 1971). Finally, E_1 and E_2 species can likely be mixed by collisions, if these involve short-range forces which induce high-order electric moments. Alternatively, mixing between E_1 and E_2 species can occur in the dipole sense as well via the $K = 0$ level, since the $K = 0$ E_1 and E_2 states are actually one and the same state. This can be expected to equilibrize E_1 and E_2 species at a fairly rapid rate.

More likely, figure 2 also shows that the observations might be explained if the $K = 2$ levels are overpopulated in Ori A and the $K = 1$ levels in Sgr B2. Such preferential population of certain K levels has already been observed for CH_3CN in Sgr B2 (Solomon *et al.* 1971); if such effects are to be explained by infrared trapping, for example, then one might hope to notice some common features in the disposition of energy levels in CH_3CN and in CH_3OH, but none are apparent.

Whether either of these two possibilities must be given serious consideration can be determined only on the basis of future observations of many additional methyl alcohol lines in both Ori A and Sgr B2 as well as better upper limits on some of the lines already observed. Calculations of level populations under the action of collisions and spontaneous decay, perhaps for a variety of assumed collisional selection rules, would also be very helpful. Litvak has made such calculations (private communication), the results of which are very strongly dependent upon the details of the collisional interactions; their relevance is not clear without observations of additional transitions.

We are grateful to J. Carter of the Haystack Observatory for the construction of the local oscillator system and for his help during the observations. We are indebted to R. W. Wilson for his invaluable assistance with the receiver. We thank Professor R. M. Lees for use of his torsional line strength calculations in advance of publication. The NRAO made available several klystrons for the observations. Research at the NEROC Haystack Observatory is supported by the National Science Foundation (grant GP-25865; contract NA59-7830).

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