Hunt for Molecules
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THE DEEP SEARCH OF O_2 IN INTERSTELLAR SPACE

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Abstract. Soon after CO was discovered in interstellar space and the first chemical models were published, predictions of large amounts of O_2, almost as much as that of CO, were made. From a simple picture where the cosmic abundance of O is about twice as much as that of C and all available carbon is locked into CO, half of the oxygen atoms should be available to form O_2 and H_2O with a large predominance for O_2 in cold environments. An O_2/CO ratio between 0.2 and 0.5 was thus predicted. However the search was obviously difficult to pursue due to the large quantities of telluric molecular oxygen. The quest actually started in 1985 and the author joined it in 1992. We retell this quest up to the most recent results where the Odin satellite with an unprecedented sensitivity may have detected the line but at a level 3 orders of magnitude below the early predictions and we give a look towards the future.

1 Introduction

When molecular studies became an important field of radioastronomy in the early '70s, the molecules containing oxygen attracted a lot of attention: OH and H_2O which had been discovered first through cm radiation in the '60s, and then CO, CH_3OH, HCO^+, H_2CO, etc. Because O is the most abundant of heavy elements (only second to H and He), the understanding of its chemistry is of paramount importance to master the difficult problem

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of chemical evolution of interstellar dark clouds. Among oxygen bearing species, early chemical models predicted large amounts of CO, O_2 and H_2O (e.g. Clavier et al. 1978). Only CO was easy to detect from the ground, the 22 GHz line of H_2O which was detected beforehand was only a maser line requiring a warm and dense environment, to be seen only near massive star formation sites and difficult to interpret in terms of water abundance. However, on top of their importance in the chemical network, these species were predicted as the main coolants in dark clouds (Goldsmith & Langer 1978) and thus it was also important to confirm this prediction which has some importance on the way dark clouds can cool down and contract to form stars.

Molecular oxygen and water being largely present in the terrestrial atmosphere and specifically in the troposphere, they cannot be easily observed from the ground. The high pressure enlarges the lines so much that it is difficult to take advantage of the astronomical sources Doppler shift velocity effects to escape them. Through the history of this quest, we will review the different strategies which have been used until now to search for O_2. After a brief summary of O_2 properties, we will recall the early quest from the ground and then from balloon experiments and finally from space.

2 O_2 quantum properties

O_2 is a homonuclear molecule with non-zero electronic spin ($^3\Sigma$ state). Spin (described by the quantum number S) and end-over-end rotation (described by N) interact to form triplet states (J = N+S), except for N = 0, where only J = 1 is allowed. O_2 has no electric dipole moment but does have a magnetic spin moment. In this case, selection rules are of two types. The first type corresponds to fine structure transitions :

$$\Delta N = 0, \Delta J = \pm 1$$

And the second type to rotational level transitions :

$$\Delta N = \pm 2, \Delta J = 0, \mp 1$$

(including spin state transitions when $\Delta J \neq 0$)

For symmetry reasons due to the homonuclearity, even values of N are forbidden for $^{16}O^{16}O$ while they are authorized for $^{16}O^{18}O$. This difference is important because it means that while the difference in frequency for the same transitions of $^{16}O^{16}O$ and $^{16}O^{18}O$ is too small to bring the rarer species outside the wings of the main species, there exists transitions for even N levels of $^{16}O^{18}O$ which have no correspondance for the main species and which can benefit from an unblocked atmosphere. For example the $(N_J : 1_1 - 1_0)$ transition lies at 118.750343 GHz for $^{16}O^{16}O$ and at 118.7599 GHz for $^{16}O^{18}O$ while the pure rotational $(N_J : 2_1 - 0_1)$ transition of $^{16}O^{18}O$ stands at 233.94618 GHz in the very well-know "clear" 1.3 mm window, close to the CO (J : 2-1) line. Fortunately, this $(N_J : 2_1 - 0_1)$ line is one of the two ground lines of $^{16}O^{18}O$ and has the strongest line strength among the unblocked lines. It makes this line the best candidate for ground-based searches of molecular oxygen in our Galaxy.

Magnetic spin moment transitions have very low strength and the resulting Einstein A coefficient (spontaneous de-excitation) is very weak, even weaker than for CO (for the $N_J : 1_1 - 1_0$ transition, $A_{ul} = 4.46 \times 10^{-9}$ s^{-1} and for the $N_J : 2_1 - 0_1$ transition of $^{16}O^{18}O$, $A_{ul} = 1.33 \times 10^{-8}$ s^{-1}, to be compared with 7.45×10^{-8} s^{-1} for the CO J : 1-0 line and 7.16×10^{-7} s^{-1} for the CO J : 2-1 line). It means that the line becomes optically thick only for large colum densities and is thus very difficult to detect. $^{16}O^{18}O$ has also an electric dipole moment, allowing $\Delta N = \pm 1$ type transitions. However, the electric dipole moment is still much weaker than the magnetic spin moment and would probably be undedectable.

3 First searches of O_2 from the ground (1985-1997)

From the ground two alternative routes exist to search for O_2. Either search for the rare isotopologue $^{16}O^{18}O$ in the Galaxy or search for the main species ($^{16}O^{16}O$) at high redshifts ($z \geq 0.02$).

3.1 Search for $^{16}O^{18}O$

As mentioned in the previous section, the $(N_J : 2_1 - 0_1)$ $^{16}O^{18}O$ line at 234 GHz is usable to search for molecular oxygen in our Galaxy. The telluric line is about 20 K in amplitude and about 20 km s^{-1} wide. By a careful choice of the sources and/or the time of the year in order to maximise the Doppler correction due to the combination of the source LSR correction and Earth rotation around the Sun, the telluric line can easily be avoided.

$^{16}O^{18}O$ having two possible sites for exchanging an ^{18}O atom with a ^{16}O atom, its relative abundance to the main species should be twice as much as the isotopic ratio ($\approx$ 500), that is $^{16}O^{16}O/^{16}O^{18}O \approx 250$. The rarity of this isotopologue leaves the atmosphere transparent enough for us to see through it at frequencies close to the rest frequency of $^{16}O^{18}O$ but it also means that in the interstellar space, the species is rather rare and the expected line very weak. A few mK were expected, certainly less than 20 mK. And the line, optically thin, was expected also to be very narrow.

When the first Schottky receivers at 1.3 mm became sensitive enough, the search began (Goldsmith et al. 1985, Liszt & van den Bout 1985) with upper limits hardly reaching the level of O_2 abundance predicted by the chemical models (Black & Smith 1984). The search was renewed when new technology receivers using the very high sensitivity SIS mixers became available. The search was conducted in Europe with the IRAM 30-m (Fuente et al. 1993) and the small 2.5-m POM-2 telescope (Pagani et al. 1993). Neither telescope was as good as present day telescopes. The main advantage of POM-2 (Castets et al. 1988) was its availability for long and deep searches which was clearly not the case for the heavily oversubscribed 30-m. 25 weeks in total were spent at POM-2, mostly by P. Maréchal and myself to search for O_2. The preliminary detection in L183 presented in Pagani et al. (1993) was not confirmed in other sources including the same source at other positions (Maréchal et al. 1997). We had spent 200 hours of integration to get that preliminary 3–4 σ detection and felt it was more efficient to search the same line elsewhere than to repeat this very difficult observation. We proved to be wrong because the line does not belong to O_2 as shown by the satellite searches later on (see below). Whether we had detected an artefact or a line of another species is not clear. POM-2 has closed down and no other telescope is ready to spend 50 hours (considering the large improvement of telescope sensitivities nowadays) to check this point. However, the main result of our search was that for the first time the upper limit on O_2 abundance was conservatively put a factor of 2 lower than the standard chemical models for dark clouds in the Galaxy (Maréchal et al. 1997). No other search has been performed since that time.

3.2 Search for redshifted O_2

In parallel to his search for $^{16}O^{18}O$ in the Galaxy, H. Liszt (1985) did the first serious search of O_2 in an external galaxy (NGC 7674, a Seyfert galaxy for which CO had been detected previously) at a redshift high enough ($z \approx 0.03$) to escape the telluric absorption. A previous attempt in front of extragalactic continuum sources had no chance to report a convincing result (Matsakis et al. 1982). Because the main isotopologue is 250 times more abundant than the rare one and despite a slightly weaker Einstein A_{ul} coefficient, the abundance limit one can get is in principle much deeper than in the Galactic searches and in the last published tentative search of O_2, Combes et al. (1997) find $O_2/CO < 0.006$ (3 σ) in a source (B0218+357 at $z = 0.685$) where they also detected H_2O (Combes & Wiklind 1997) and several other species. Of course, they have little information on what kind of clouds they have on their line of sight, with which filling factor, and what are the physical conditions of these clouds, possibly not suitable to hold large amounts of O_2, but the detection of many other species and not only CO seems a strong indication that this limit is indeed meaningful.

4 Leaving the ground (1997–)

To be able to observe the main O_2 species in our Galaxy, one has to go high enough in the atmosphere where the telluric lines are narrow enough to escape them with sources only a few tens of km s^{-1} away in velocity. Airplanes like the KAO or the future SOFIA flying at 12–14 km altitude are not high enough to do that but balloons flying close to 40 km altitude reach a pressure level of a few mbars only and the telluric lines, still opaque in their centers, are now narrow enough to allow observations of water or molecular oxygen. Finally, satellites are free from any constraint and can observe the sky for several years.

4.1 Balloons are not so easy to use

Two teams tried to develope a balloon-borne radiotelescope tuned to O_2 frequencies. One team at UCSB made two attempts (1993 and 1994) with a 119 GHz SIS receiver but both flights failed for technical reasons and no result has been obtained. The other team, originally completely French prepared an experiment to measure both H_2O @ 380 GHz and O_2 @ 368 GHz with a large telescope (2-m) to be carried on a balloon. This was the largest pointed gondola ever launched (PRONAOS). Many technical difficulties delayed the project and eventually the team moved to another gondola which had already flown, PIROG, and which was constructed and ran by a Swedish team. The telescope was much smaller (60 cm) but ready to fly. We took that opportunity to move from the $(N_J : 3_2 - 1_2)$ 368 GHz to the $(N_J : 3_2 - 1_1)$ 424 GHz line combining it with the (J :4–3) ^{13}CO transition. This small frequency difference, technically marginal, allowed to gain a factor of 10 in sensitivity as the $(N_J : 3_2 - 1_1)$ Einstein coefficient of spontaneous emission is 10 times larger than for the $(N_J : 3_2 - 1_2)$ line (2.40 $\times 10^{-8}$ and 1.92 $\times 10^{-9}$, respectively). The balloon, carrying PIROG-8 was succesfully launched from Aire-sur-l'Adour, south of France, and no O_2 was found in our Galaxy. Two sources were looked at, NGC 7538 and W51, possibly not the best ones but the choice of sources was extremely limited due to technical limitations on the gondola and the necessity to escape the residual telluric line. We improved by a factor of 2 the upper limit we had obtained with POM-2 ($O_2/CO < 0.04$, Olofsson et al. 1998).

4.2 SWAS and ODIN

Finally came the era of the satellite missions. SWAS was equipped with two submm receivers, one tuned to the $(N_J : 3_3 - 1_2)$ line of O_2 at 487 GHz. The main dish (54 × 68 cm), the Schottky receiver ($T_{sys} \approx 5000$ K) and the low velocity resolution (0.6 km s^{-1}) were representing some limitations (especially in dark cloud cores). These limitations were partly compensated by long integrations and a large choice of sources. Eventually upper limits of $O_2/H_2 = 3 \times 10^{-7}$ were achieved (Goldsmith et al. 2000), representing a O_2/CO ratio of 0.003, a factor of 10 improvement on the PIROG 8 experiment.

Odin, still active, is an improved version of SWAS. The main dish is twice as large (1.1 m diameter), receivers are more sensitive (3000 K typically), tunable in a large range, with selectable spectral resolution and most important, a dedicated receiver for O_2 at 119 GHz (the ground transition) with a very low noise (600 K) has been added. Its main disadvantage is that the angular resolution is only $9'$. However, this is the ideal instrument to search for O_2 as this line is stronger than the 487 GHz line in any source below 100 K, and especially in dark clouds where the gain in sensitivity is of almost two orders of magnitude. We had no more success than SWAS to detect O_2, reporting upper limits down to $O_2/H_2 = 7\text{–}8 \times 10^{-8}$ (Pagani et al. 2003).

Difficulties have arisen to push the limit further down because the receiver phase-lock broke down and its remaining stability is only of the order of 1 MHz. Current work is being done to try to reduce these unlocked data and estimate precisely the limitation of sensitivity due to the failing phase-lock. Upper limits or marginal detections to the level of 2–3 $\times 10^{-8}$ could be obtained.

5 Discussion and conclusion

The absence of O_2 in the interstellar medium came as a big surprise and is still a puzzle to astrochemists despite the fact that several explanations have been advocated (either a naturally low abundance due to e.g. sticking of O_2 on the grains, Vandenbussche et al. 1999, or larger C/O ratio than the solar one, Roberts & Herbst 2002, which can be due to different reasons like sticking of O on the grains to form H_2O, Bergin et al. 2000, or a more efficient destruction process due to e.g. deeper penetration of UV rays, Casu et al. 2001, turbulence taking surface layer C^+ deep inside the cloud, Chièze & Pineau des Forêts 1989, higher HI abundance inside clouds, Willacy et al. 2002, etc).

Though the importance of O_2 as a main coolant of dark clouds, to lead to contraction and protostar formation, is now abandoned, it is still useful to try to detect it in a variety of conditions because this will bring a strong constraint on astrochemical models but also some hints at what makes O_2 so rare. Is it possible to do so ? Odin is hampered by its failing phase-lock and the next space mission, the Herschel Space Observatory will have only a 487 GHz receiver and little time to spend on this particular line. Its advantages upon SWAS and Odin is a larger dish (3.5 m) and a very sensitive receiver (200 K can be expected). If a compact O_2 source of a few tens of mK exists somewhere it will be easily detected by Herschel but low spread emission or low abundance even in the densest cores will not. It is thus not certain that Herschel will bring the light on the O_2 mystery...

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