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*Letter to the Editor***Constraints on the abundance of solid O₂ in dense clouds from ISO-SWS and ground-based observations[★]****B. Vandenbussche¹, P. Ehrenfreund², A.C.A. Boogert⁴, E.F. van Dishoeck^{2,3}, W.A. Schutte², P.A. Gerakines⁵, J. Chiar⁷, A.G.G.M. Tielens⁴, J. Keane⁴, D.C.B. Whittet⁵, M. Breitfellner⁶, and M. Burgdorf⁶**¹ Instituut voor Sterrenkunde, K.U.Leuven, Celestijnenlaan 200B, B-3001 Heverlee, Belgium² Raymond and Beverly Sackler Laboratory for Astrophysics at Leiden Observatory, 2300 RA Leiden, The Netherlands³ Leiden Observatory, P.O. Box 9513, 2300 RA Leiden, The Netherlands⁴ Kapteyn Astronomical Institute, P.O. Box 800, 9700 AV Groningen, The Netherlands⁵ Rensselaer Polytechnic Institute, Dept of Physics, Applied Physics & Astronomy, Troy, NY 12180, USA⁶ ISO Data Centre, Astrophysics Division, Space Science Department of ESA, Villafranca, P.O. Box 50727, E-28080 Madrid, Spain⁷ NASA-Ames Research Center, Mail Stop 245-3, Moffet Field, CA 94035, USA

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Abstract. The Short Wavelength Spectrometer (SWS) on-board the Infrared Space Observatory (ISO) has been used to search for solid O₂ in cold dense clouds at 6.45 μm . Additional constraints on the O₂ abundance are obtained from analysis of the 4.67 μm solid CO absorption profile observed from the ground. We derive upper limits of 50% and 100% of solid O₂ relative to solid CO toward the protostellar sources R CrA IRS2 and NGC 7538 IRS9 respectively, corresponding to abundances of 30×10^{-6} and 15×10^{-6} relative to n_H . These results indicate that the abundance of solid O₂ in dense clouds accounts for less than 6% of the total oxygen budget in the interstellar medium. The reservoirs of oxygen in dense clouds are discussed, taking into account recent measurements of oxygen-bearing species.

Key words: ISM: abundances – ISM: molecules – ISM: dust, extinction – infrared: ISM: lines and bands

1. Introduction

Oxygen, which is cosmically the most abundant element after H and He, plays an important role in interstellar chemistry and in the energy balance of interstellar clouds. Knowledge of its major reservoirs, both in the gas and on grains, is therefore essential. The principal oxygen-bearing species in diffuse and dense clouds have been the subject of considerable discussion (see van Dishoeck & Blake 1998 for a review). In some regions, up to 50% of the oxygen is unaccounted for if solar abundances are assumed.

Send offprint requests to: B. Vandenbussche

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Recently, a revised O⁰ abundance has been determined for the local diffuse interstellar medium using the Hubble Space Telescope. Measurements of the OI] line at 1356 Å toward several stars indicate gaseous atomic oxygen abundances of 319 ± 14 per $10^6 n_H$ (Meyer et al. 1998). Together with an estimated abundance of O in silicates of 180 per $10^6 n_H$, this comes close to the oxygen abundance in B stars. At the same time, ground-based, balloon-borne and ISO satellite data yielded abundances of several other oxygen-bearing species, in both the gas phase (including O⁰, O₂ and H₂O) and the solid state (including H₂O, CO₂ and CO) (see discussion in Sect. 4). A re-evaluation of the oxygen budget in dense clouds is therefore warranted.

Theoretical models predict that oxygen could be accreted onto grains from the gas in the form of solid O₂ (Tielens & Hagen 1982), mixed with CO ice in apolar ices. O₂ is an infrared inactive molecule, which does not have any signature in the infrared and radio range and is therefore difficult to observe. Different methods to detect solid O₂ on interstellar grains have been discussed by Ehrenfreund & van Dishoeck (1998), and the most direct opportunity is the search for the weak fundamental O₂ transition at 6.45 μm (Ehrenfreund et al. 1992). In the solid state, this transition becomes weakly infrared active due to interactions with neighboring atoms, and laboratory results indicate that the band strength of molecular oxygen depends on the ice matrix. Other constraints on the O₂ abundance come from the analyses of solid CO profiles and from searches for photoproducts of O₂ (such as O₃, CO₃, etc.). Strazzulla et al. (1997) searched for a means of indirectly detecting O₂ and N₂ through changes induced in the CO absorption profile as a result of ion irradiation and the products formed during the radiolysis. Here, we present new ISO-SWS and ground based data in order to constrain the solid O₂ abundance in dense clouds.

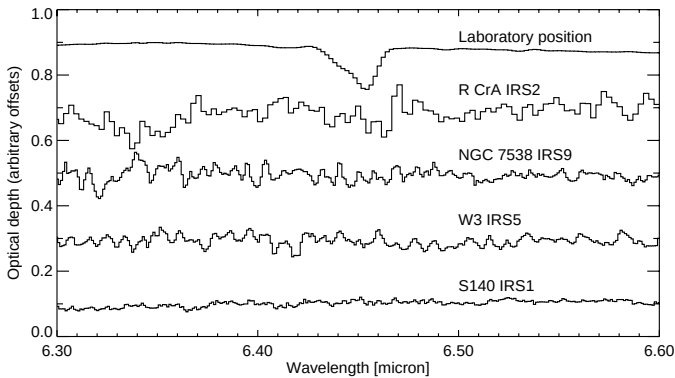


Fig. 1. ISO-SWS spectra of R CrA IRS2, NGC 7538 IRS9, W3 IRS5 and S140 IRS1 between 6.3 and 6.6 μm compared to that of solid O₂ in the laboratory, whose fundamental transition falls at 6.45 μm .

2. Observations

Because of the low evaporation temperatures of apolar ices (~ 20 K), their detections in the interstellar medium are limited to regions which contain a large amount of cold, quiescent material, far away from protostars. Our search for solid O₂ was therefore directed toward lines of sight with large apolar CO ice abundances. Chiar et al. (1998) have recently summarized solid CO observations for a large number of regions. On the basis of their Fig. 7, two lines of sight stand out as excellent candidates: R CrA IRS2 and NGC 7538 IRS9. R CrA is located in the Corona Australis complex, which, like Taurus, appears cold in nature. Toward R CrA IRS2 the amount of apolar CO ice is more than 50% that of water ice, the highest solid CO:H₂O ratio ever measured (Chiar et al. 1998). Toward NGC 7538 IRS9, the abundance of apolar CO ice is 19% relative to water ice (Chiar et al. 1998). Gas and solid-state measurements indicate a cold cloud component (Mitchell et al. 1990, Chiar et al. 1998), where solid O₂ may also be present in abundance. We also present the ISO-SWS observations of two deeply embedded protostellar objects: W3 IRS5 and S140 IRS1. The higher temperature of these clouds is not favorable for the presence of solid O₂, but both sources have high fluxes in the 6.2–6.6 μm region, yielding higher signal-to-noise in their ISO spectra.

The ISO-SWS observations of these sources were performed in the AOT6 observation mode. The integration times in the 6.2–6.6 μm region were 2400 s for R CrA IRS2 and 750 s for NGC 7538 IRS9, W3 IRS5 and S140 IRS1. The data were reduced with the SWS Interactive Analysis package (de Graauw et al. 1996a). The standard product generation steps were done, with manual dark current subtraction and exclusion of data points that suffered from uncorrected events in the read-out electronics (visible as signal jumps) to obtain the best S/N possible.

The SWS spectra have been divided by the local continuum level around 6.45 μm . The low value of S/N = 10 in the final spectrum of R CrA IRS2 (Fig. 1) is due to the low brightness of the source (5.5 Jy at 6.45 μm). At these flux levels the detector noise and signal drifts dominate the S/N of SWS observations in this spectral region. The S/N ratio is 20 in the final spectra

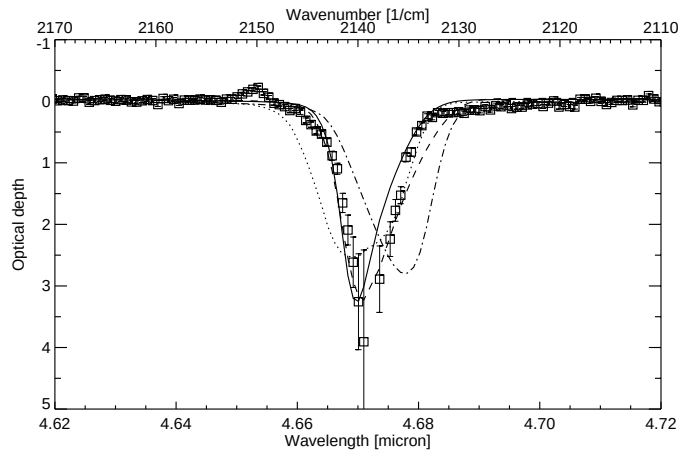


Fig. 2. The CO band of R CrA IRS2 observed with the cooled grating spectrometer CGS4 on the United Kingdom Infrared Telescope (boxes with error bars) compared to laboratory spectra of CO:O₂=10:5 (solid line), CO:O₂=10:7 (dashes), CO:O₂=1:1 (dot-dashes) and CO:O₂:CO₂:N₂=100:500:50:100 (dots).

of NGC 7538 IRS9, W3 IRS5 and S140 IRS1. NGC 7538 IRS9 is brighter than R CrA IRS2 around 6.45 μm (33 Jy), but here the spectrum is dominated by the long-wavelength wing of the solid H₂O bending mode located at 6.0 μm .

3. Results

From the spectra displayed in Fig. 1, we derive the following 2σ upper limits on the optical depth of the fundamental transition of solid O₂ at 6.45 μm : 0.2 for R CrA IRS 2 and 0.1 for NGC 7538 IRS9, W3 IRS5 and S140 IRS1.

The integrated cross section of solid O₂ has been previously estimated as 1×10^{-19} cm/molecule in multicomponent mixtures (Ehrenfreund et al. 1992). New measurements indicate that the cross section in apolar ices can be as small as 5×10^{-21} cm/molecule. To calculate the upper limits on the column density of solid O₂ we adopt a cross section of 1×10^{-20} cm (measured for a CO:O₂=2:1 ice mixture). The upper limits on O₂ column densities are thus 1×10^{20} cm⁻² for R CrA IRS2 and 5×10^{19} cm⁻² for NGC 7538 IRS9, W3 IRS5 and S140 IRS1.

Another method of constraining the abundance of solid O₂ is the analysis of the solid CO absorption profile. Fig. 2 shows the CO band toward R CrA IRS2 as observed with the cooled grating spectrometer CGS4 on the United Kingdom Infrared Telescope (Chiar et al. 1998). Saturation of the CO band results in large error bars near its peak position. Toward R CrA IRS2, the CO band is very narrow, FWHM = 3.2 cm⁻¹. Its profile is consistent with pure CO (Chiar et al. 1998) and ice mixtures of O₂: CO = 1:2 or O₂:CO = 20:1 (Ehrenfreund et al. 1997, Elsila et al. 1997). However, at very large quantities of O₂ the CO feature shows a distinctive redshift to 2135.8 cm⁻¹, which is inconsistent with the observations of R CrA IRS2. In a more complex mixture of CO:O₂:CO₂:N₂ = 100:500: 50:100 (Elsila et al. 1997) the position falls close (2141.6 cm⁻¹) but the width of the profile (>6.8 cm⁻¹) is inconsistent with the observa-

Table 1. Column densities and abundances per 10⁶ n_H of oxygen bearing species in R CrA IRS2 and NGC 7538 IRS9 compared to the O⁰ abundance in the diffuse ISM

Species	R CrA IRS2		NGC 7538 IRS9	
	column density (cm ⁻²)	O Abundance (/ 10 ⁶ n _H)	column density (cm ⁻²)	O Abundance (/ 10 ⁶ n _H)
CO ice	1.2±0.3 × 10 ¹⁸ (c)	30.0±7.5	1.2±0.1 × 10 ¹⁸ (c)	7.5±0.6
CO gas	1.6±0.2 × 10 ¹⁸ (e)	40.0±3.7	1.2 × 10 ¹⁹ (f)	75
H ₂ O ice	2.1 × 10 ¹⁸ (a)	52.5	8.0 × 10 ¹⁸ (b)	50
H ₂ O gas	< 10 ¹⁸	< 25	< 3.0 × 10 ¹⁷ (g)	< 1.9
CO ₂ ice	8.5±1.7 × 10 ¹⁷ (k)	42.7±8.5	16.3±1.8 × 10 ¹⁷ (j)	20.4±2.3
OCN ⁻			3.3 × 10 ¹⁶ (h)	0.2
CH ₃ OH			4.8 × 10 ¹⁷ (b)	3
HCOOH			2.4 × 10 ¹⁷ (i)	3
O ₂ gas	< 2.6 × 10 ¹⁷ (m)	< 12.9	< 4.8 × 10 ¹⁷ (n)	< 6
O ₂ ice	< 6.0±1.5 × 10 ¹⁷	< 30.0±7.2	< 1.2±0.5 × 10 ¹⁸	< 15.0±0.6
H gas	4.0 × 10 ²²		1.6 × 10 ²³	
Total		< 233.1±26.9		< 182.5±3.5
O ⁰ diffuse medium		319±14		319±14
Deficit		> 85.9±40.9		> 137.5±17.5

References: (a) Tanaka et al. 1994; (b) Allamandola et al. 1992; (c) Chiar et al. 1998; (e) Harju et al. 1993; (f) Mitchell et al. 1990; (g) van Dishoeck & Helmich 1996; (h) Keane & Schutte, in preparation; (i) Schutte et al. 1996; (j) Gerakines et al. 1999; (k) Boogert et al. 1999; (m) Marechal et al. 1997; (n) Olofsson et al. 1998

tions, see Fig. 2. An abundance of solid O₂ which exceeds the abundance of solid CO is not favored by theoretical models, taking into account recent values for the abundance of gas-phase oxygen and carbon (Tielens & Hagen 1982, Meyer et al. 1998, Cardelli et al. 1996). From the data presented in Fig. 2 we conclude that a mixture with O₂/CO = 70% results in a band wider than the observed CO profile of R CrA IRS2. Furthermore, the position of the CO band profile is inconsistent with a mixture CO:O₂=1:1. We thus derive an upper limit of 50% O₂ ice with respect to CO toward R CrA IRS2.

Elsila et al. (1997) provided a reasonable fit in band position (2142 cm⁻¹) to the CO band of NGC 7538 IRS9 observed by Tielens et al. (1991) with a CO:O₂:CO₂:N₂ = 100:500:50:100 mixture. However, the fit was done in transmittance. In optical depth it is apparent that the bandwidth of the mixture (FWHM=7.1 cm⁻¹) is much larger than the observed bandwidth (FWHM=4.75 cm⁻¹). The difference is caused by saturation effects owing to the large optical depth of the interstellar CO feature. Chiar et al. (1998) used a two-component fit without any solid O₂ to match the profile of the CO band in NGC 7538 IRS9. Additional measurements listed in their Table 5 show that a number of good fits to the apolar CO component can be achieved for O₂-rich mixtures where O₂/CO = 50–100%. We therefore adopt an upper limit of O₂/CO=1, which is still consistent with theoretical models. This abundance translates to 19% O₂ relative to H₂O ice toward this source, and accounts for less than < 3% of the total interstellar oxygen budget. Due to the larger width of the CO band toward NGC 7538 IRS9 (4.75 cm⁻¹), the presence of O₂ ice can not be as well constrained as for R CrA IRS2.

4. Discussion

Table 1 lists the column densities of important oxygen-bearing species and the total hydrogen column density (H and H₂) in R CrA IRS2 and NGC 7538 IRS9. The hydrogen column density of the interstellar medium is often derived from the visual extinction. In dense clouds, the grain sizes are known to be larger than in the diffuse medium so that the amount of visual extinction per unit mass is larger than in the diffuse medium. Observations of the field star Elias 16 behind the Taurus dense cloud show a A_v/τ(9.7) ratio of 31.2, as compared to 18.5 for the diffuse medium towards CygOB#12 (Whittet et al. 1988, Whittet et al. 1997). Since the grain sizes in R CrA IRS2 are expected to be comparable, we assume that the same factor of 1.69 times the A_v/N_H ratio in the diffuse ISM can be assumed. With an A_v value of 35 (Chiar et al. 1998), we thus derive a total H column density of 4.0 × 10²² cm⁻² for R CrA IRS2. The A_v of NGC 7538 IRS9 derived from the depth of the 9.7 μm feature (Willner et al. 1982) is insensitive to the grain size. The inferred column density is 1.6 × 10²³ cm⁻² (Chiar et al. 1998).

The column density of CO gas of R CrA IRS2 is based on the C¹⁸O 1-0 data by Harju et al. (1993). We derive N(C¹⁸O)=3.1 × 10¹⁵ cm⁻² for typical dark cloud conditions. For a normal ¹⁶O/¹⁸O ratio of 500, this implies a column density of gaseous CO of 1.6±0.2 × 10¹⁸ cm⁻². The CO gas column density towards NGC 7538 IRS9 was determined by Mitchell et al. (1990) from the ¹³CO IR absorption. From the ISO-SWS spectrum of R CrA IRS2 we determined an upper limit of 10¹⁸ cm² for hot H₂O, assuming a line width > 3 km s⁻¹. The upper limit on the H₂O gas column density in NGC 7538 comes from the ISO spectra. The column densities of the CO

and H₂O ice are determined from ground based spectra. The ISO spectrum of NGC 7538 IRS9 also revealed other oxygen bearing ice species like OCN⁻ and HCOOH while the presence of CH₃OH was known from ground based spectra.

The recent balloon experiment PIROG 8 searched for the 425 GHz gas-phase O₂ line toward NGC 7538 IRS9 and W51 (Olofsson et al. 1998). No emission could be detected, and the inferred upper limits on the O₂/CO ratio are 0.04 and 0.05 (3 σ) for these regions. This value leads to an upper limit on the O₂ gas column density in NGC 7538 IRS9 of $6 \times 10^{16} \text{ cm}^{-2}$. Marechal et al. (1997) searched for gas-phase ¹⁶O¹⁸O in dark clouds. For regions in L134 which are comparable to R CrA IRS2, they give an upper limit of O₂/CO < 0.15, which would correspond to an O₂ upper limit of $2.6 \times 10^{17} \text{ cm}^{-2}$ for R CrA IRS2.

The upper limits on solid O₂ from the CO profile deconvolution found in this paper are $6.0 \pm 1.5 \times 10^{17} \text{ cm}^{-2}$ in R CrA IRS2 and $1.2 \pm 0.5 \times 10^{18} \text{ cm}^{-2}$ in NGC 7538 IRS9. These limits are also consistent with those derived from the 6.45 μm feature. Table 1 contains the abundances of oxygen bearing species in R CrA IRS2 and NGC 7538 IRS9. We compare these values with the abundance of gaseous O⁰ in the diffuse ISM (Meyer et al. 1998), which is the oxygen available to form these species in dense clouds. Not listed are the dust species (silicates, oxides) which are assumed to contain the same amount of oxygen in both diffuse and dense clouds. We estimate that in R CrA IRS2 at least 27% ($\pm 13\%$) of the oxygen is unaccounted for. More significant is that at least 43% ($\pm 6\%$) of the oxygen seems to be missing in NGC 7538 IRS9. Recent Kuiper Airborne Observatory and ISO observations of the [OI] 63 μm line indicate that up to 40% of the oxygen could be in atomic form (Poglitsch et al. 1996, Baluteau et al. 1997). However, these observations trace only the foreground material and not the dense cloud. Some of the missing oxygen in the two protostellar objects discussed here might be in atomic oxygen, but further observational evidence is needed.

5. Conclusions

In summary, a search for solid O₂ toward the most promising cold clouds with ISO-SWS and ground-based observations has led to an upper limit on the amount of condensable oxygen found in the form of O₂ ice of 10%. Recent observations with the sub-mm satellite SWAS (Submillimeter Wave Astronomy Satellite) have confirmed the low abundance of gaseous O₂ in cold clouds, such as NGC 7538 IRS9. Furthermore, the amount of oxygen found in “cold” H₂O toward such targets is negligible.

The low abundances of solid and gaseous O₂ suggest that O⁰ should be the dominant form of oxygen in dense clouds. Further measurements with SWAS using longer integration times and future space missions will give more conclusive evidence on the abundance of gas phase O₂ in interstellar clouds.

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