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Constraining the ortho-to-para ratio of H_2 with anomalous H_2CO absorption⋆

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ABSTRACT

Context. The ortho-to-para ratio (OPR) of molecular hydrogen is a fundamental parameter in understanding the physics and chemistry of molecular clouds. In dark and cold regions, however, H_2 is not directly observable and the OPR of H_2 in these sources has so far remained elusive.
Aims. We show that the 6 cm absorption line of ortho-formaldehyde (H_2CO) can be employed to constrain both the density and the OPR of H_2 in dark clouds.
Methods. Green Bank Telescope (GBT) observations of ortho-H_2CO toward the molecular cloud Barnard 68 (B68) are reported. Non-LTE radiative transfer calculations combined with the well-constrained structure of B68 are then employed to derive the physical conditions in the absorption region.
Results. We provide the first firm confirmation of the Townes & Cheung mechanism: propensity rules for the collisions of H_2CO with H_2 molecules are responsible for the sub-2.7 K cooling of the 6 cm doublet. Non-LTE calculations show that in the absorption region of B68, the kinetic temperature is ∼10 K, the ortho-H_2CO column density amounts to $\sim 2.2 \times 10^{13}$ cm^{-2}, the H_2 density is in the range $1.4–2.4 \times 10^4$ cm^{-3}, and the OPR of H_2 is close to zero. Our observations thus provide fresh evidence that H_2 is mostly in its para form in the cold gas, as expected from theoretical considerations. Our results also suggest that formaldehyde absorption originates in the edge of B68, at visual extinctions $A_V \lesssim 0.5$ mag.

Key words. astrochemistry – ISM: molecules – ISM: abundances – ISM: individual objects: Barnard 68

1. Introduction

The ortho-to-para ratio (OPR) of molecular hydrogen (H_2) is a fundamental parameter in understanding the physics and chemistry of interstellar molecular clouds. Chemical models have shown, in particular, that the degree of deuteration of molecular ions and molecules during the pre-protostellar collapse is very sensitive to the form of H_2 (Flower et al. 2006; Pagani et al. 2009). In these environments, H_2 forms on grain surfaces, and it is generally assumed that the initial OPR is equal to the statistical value of 3. Although the details of the H_2 formation mechanism remain uncertain, it is also commonly accepted that ortho-to-para conversion of H_2 occurs subsequently in the gas phase by means of proton exchange reactions with H^+ and H_3^+. Although the timescales associated with these reactions are large, the OPR of H_2 is expected to decrease toward zero (the Boltzmann equilibrium value at 10 K is 3.6×10^{-7}) during the lifetime of a molecular cloud and it is, therefore, a potentially valuable probe of the age of the cloud. This ratio is however expected to be not fully thermalized in dark clouds because of the recycling of hydrogen via gas phase reactions and catalysis on grains (Le Bourlot 1991). The chemical modelling of Flower et al. (2006) for $n_H = 10^4$ cm^{-3} and $T = 10$ K thus suggests that the OPR is $\sim 10^{-3}$ at steady-state, that is exceeds the Boltzmann equilibrium value by 4 orders of magnitude. In addition, in the warm gas associated with molecular shocks ($T_{kin} > 300$ K), direct observations of H_2 rovibrational lines have shown that the OPR is generally not equal to the Boltzmann equilibrium value of 3, but lies in the range 0.5−2 (e.g., Neufeld et al. 1998; Lefloch et al. 2003; Maret et al. 2009, and references therein). These relatively low values are interpreted as the legacy of earlier stages of the thermal history of the gas. There is therefore evidence, both observational and theoretical, that H_2 is mostly in its para form in cold and dark progenitor molecular clouds. However, since the gas temperature in this latter type of clouds is too low to populate excited levels of H_2, a direct measure of the OPR of cold H_2 has remained so far elusive.

The OPR of H_2 is also important for collisional excitation processes at low temperature because the interaction potential is different for ground state para-H_2 ($J = 0$) and ortho-H_2 ($J = 1$). This is because the permanent quadrupole moment of H_2 vanishes when H_2 is in its spherically symmetric ground para state ($J = 0$). As a result, rate coefficients with ortho-H_2($J = 1$) are generally higher than those with para-H_2($J = 0$). Collisional propensity rules can also be different for the two forms of H_2, as shown for example by Flower et al. (1990) in the case of ammonia and, more recently, by Troscompt et al. (2009) in the case of formaldehyde (H_2CO). One particularly interesting observation of (ortho) H_2CO is the absorption of the 6 cm line ($1_{10} \leftarrow 1_{11}$) against the 2.7 K Cosmic Microwave Background (CMB). This "anomalous" absorption, which implies an excitation temperature $T_{ex} < 2.7$ K for the doublet, was first discovered by Palmer et al. (1969) in the direction of dark nebulae. Numerous

⋆ This work has been inspired by our colleague and friend Pierre Valiron, who passed away in August 2008. This paper is dedicated to his memory.

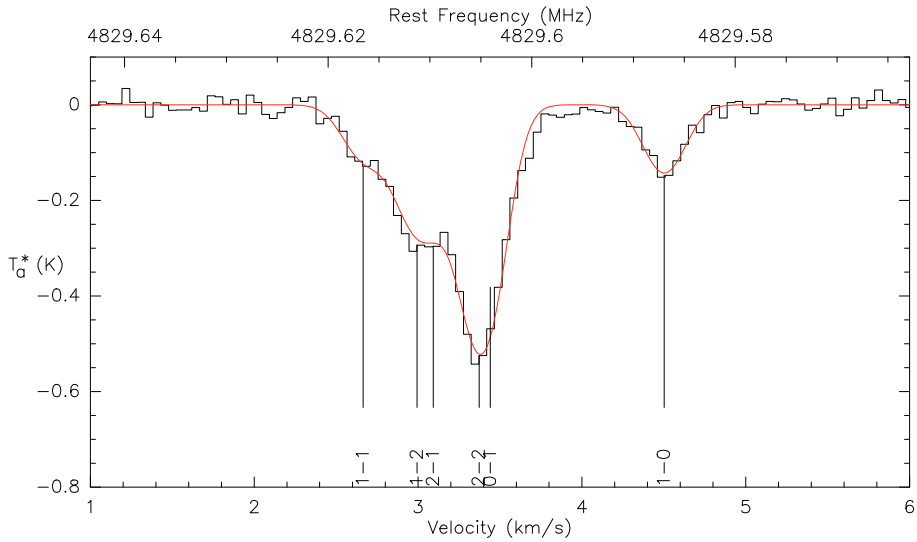

Fig. 1. Spectrum of the $1_{10} \leftarrow 1_{11}$ absorption line of ortho-H_2CO at 4.82966 GHz toward B68. The GBT spectrum is given by the black line while the red line denotes the CLASS HFS fit. The hyperfine transitions 1_{10} F′ ← 1_{11} F are marked at their respective frequencies.

theoretical studies (in particular Townes & Cheung 1969; Evans et al. 1975; Garrison et al. 1976) proposed an explanation in terms of competing radiative and collisional population transfer mechanisms. The H_2CO absorption was thus found to provide a sensitive probe of the temperature and density conditions in dark sources. All previous studies were however hampered by i) the lack of accurate collisional rates between H_2CO and H_2 (He atoms were employed as substitutes for H_2 molecules) and ii) low spectral resolution, which did not allow to properly resolve the hyperfine structure.

In this study, we present the first high spectral resolution observation of the H_2CO 6 cm absorption line toward Barnard 68 (hereafter B68), the prototype of a dense molecular cloud (Burkert & Alves 2009, and references therein). Non-local thermodynamic equilibrium (LTE) radiative transfer calculations based on the theoretical collisional rates of Troscompt et al. (2009) are then employed to derive the physical conditions in the absorption region, including constraints on the OPR of H_2 in this source. The observations and data reduction are described in Sect. 2. Radiative transfer modelling is presented in Sect. 3, and results are discussed in Sect. 4.

2. Observations

We observed the $1_{10} \leftarrow 1_{11}$ transition of ortho-H_2CO ($\nu = 4.82966$ GHz) towards B68 (RA = $17^h22^m38.2^s$, $\delta = -23°49'34''$ [J2000]) using the NRAO Robert C. Byrd Green Bank Telescope (GBT). The observation was performed on October 17, 2008. The C-band (3.95−6.1 GHz) receiver was used together with the GBT auto-correlator in nine-level sampling mode, providing a spectral resolution of 762.9 Hz (0.047 km s^{-1}). At the observed frequency, the HPBW of the GBT is 2.5′ and the main beam efficiency is $\eta_{mb} = 0.97$. The observations were performed in frequency switching mode, with a frequency throw of 100 MHz. Finally, the observation is an average of 4 × 10 min scans, for a total integration time of 40 min.

The data was reduced with the CLASS software package[1]. A first order polynomial baseline was subtracted from the spectrum, which is shown in Fig. 1. Because of the high resolution and high signal-to-noise ratio of the spectrum, several hyperfine components are resolved. Especially, the $F = 1-1$ and the (blended) $F = 1-2$ and $2-1$ components are clearly distinguishable from the (blended) $F = 2-2$ and $0-1$ components. The $F = 1-0$ component is also easily resolved. The line parameters were fitted with the HFS method in CLASS, which allows the simultaneous fitting of all the hyperfine components of a rotational transition, based on the assumption that the excitation temperature is the same for all hyperfine components and the lines do not overlap. We note that the frequencies and relative intensities of hyperfine components were taken from the laboratory measurements of Tucker et al. (1971). As shown in Fig. 1, an excellent fit to the observed spectrum is obtained, with a rms of $\sim 10^{-2}$ K.

We derived an antenna temperature T_a^* of the main hyperfine component ($F = 2-2$) of -0.44 ± 0.04 K, a source velocity $v_{LSR} = 3.374 \pm 0.003$ km s^{-1}, a total opacity of the transition $\tau_{tot} = 1.29 \pm 0.19$, and a line width $\Delta v = 0.294 \pm 0.007$ km s^{-1}. The excitation temperature was then derived from T_a^* within the Rayleigh-Jeans approximation (e.g., Martin & Barrett 1978) by assuming no clumping, that is a beam dilution factor of 1 since the spatial extent of B68 (2′) and the beam extension (2.5′) are similar. We obtained $T_{ex} = 1.63^{+0.19}_{-0.25}$ K. The present excitation temperature and opacity are thus in good agreement with those obtained by Heiles (1973), who found an average excitation temperature of 1.6 K and total opacities of ∼1 for a sample of dark clouds.

3. Non-LTE modelling

The collisional pumping mechanism invoked to explain the anomalous absorption of H_2CO was first proposed by Townes & Cheung (1969). It was based on a classical formulation showing that collisions with neutral particles should favor excitation to the lower level of each doublet of ortho-H_2CO. This collisional propensity rule, combined with the dipolar radiative selection rules, leads to an overpopulation of the 1_{11} level and a cooling of the excitation temperature of the $1_{10} \leftarrow 1_{11}$ transition below 2.7 K[2]. This scheme was subsequently supported by semi-classical and quantum calculations (Garrison et al. 1976).

[1] GILDAS package, http://www.iram.fr/IRAMFR/GILDAS

[2] This phenomenon is also observed for the $2_{11} \leftarrow 2_{12}$ transition of ortho-H_2CO at 2 cm.

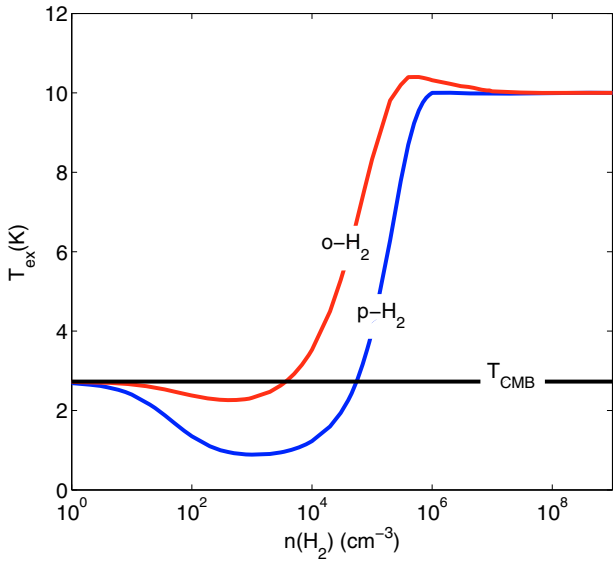

Fig. 2. Excitation temperature for the $1_{10} \leftarrow 1_{11}$ transition as a function of the H_2 density. The blue and red lines correspond to para-H_2 ($J = 0$) and ortho-H_2 ($J = 1$) as collider, respectively. The black horizontal solid line denotes the background temperature $T_{CMB} = 2.7$ K. See text for details.

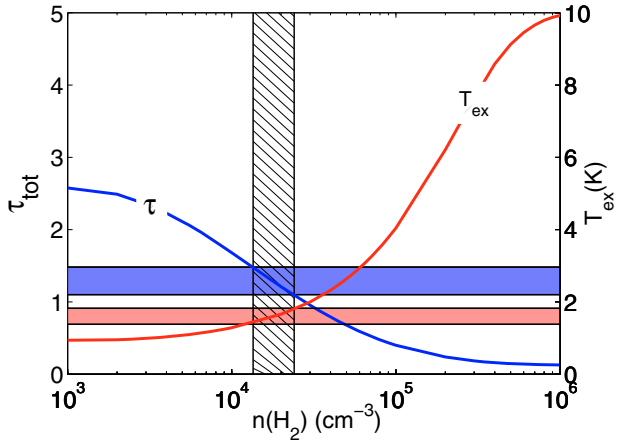

Fig. 3. Total opacity, τ_{tot}, and excitation temperature, T_{ex}, as a function of density. The blue and red shaded area denote respectively the range of observed τ_{tot} and T_{ex}. The blue and red lines denote the results of the present model for a kinetic temperature of 10 K, an ortho-H_2CO column density of 2.2×10^{13} cm^{-2} and an OPR of H_2 of zero. The dashed area indicates the density range where both τ_{tot} and T_{ex} are reproduced.

However, as mentioned in the introduction, He atoms were employed as substitutes for H_2 in all previous studies. Troscompt et al. (2009) showed that the propensity rule is weaker for H_2 than for He and, furthermore, that it depends significantly on the ortho/para form of H_2.

This is clearly illustrated in Fig. 2, where we plot the results of a non-LTE computation based on collisional excitation by either para-H_2 ($J = 0$) and He atoms or ortho-H_2 ($J = 1$) and He atoms. We note, however, that excitation by H_2 is entirely predominant, that is the contribution of He atoms was found to be negligible. Excitation by the far less abundant free electrons was neglected[3]. The hyperfine structure of H_2CO levels was neglected in the calculations (see below). We employed the MOLPOP program, written by Moshe Elitzur and Philip Lockett, to solve the radiative transfer equations using the escape probability method for a homogeneous slab (Krolik & McKee 1978). The 10 first levels of ortho-H_2CO were considered in the computation with the following basic parameters: the ortho-H_2CO column density was fixed to $N(H_2CO) = 1.0 \times 10^{13}$ cm^{-2}, the kinetic temperature to $T_{kin} = 10$ K, and the Doppler linewidth to 1 km s^{-1}. The 2.7 K CMB radiation field was obviously included but all other possible sources of radiation were neglected. The collisional data was those of Troscompt et al. (2009) and Green (1991) for excitation by H_2 and He, respectively. It is first observed, in agreement with previous calculations based on He as a collider, that cooling of the 6 cm doublet below 2.7 K does occur for densities lower than a few 10^4 cm^{-3}. This provides the first robust confirmation of the Townes & Cheung mechanism in the case of H_2 as a collider. Secondly, it is noticed that the cooling is significantly stronger for para-H_2 ($J = 0$) than for ortho-H_2 ($J = 1$), and that it occurs over a wider range of density. Hence, the cooling of the doublet is expected to depend markedly on the OPR of H_2, with diminishing intensity of the 6 cm absorption line as the OPR increases.

[3] We note that electron excitation of H_2CO can play an important role at high electron fraction ($x_e \gtrsim 10^{-5}$), in particular in diffuse clouds where electron collisions are expected to cancel the anomalous 6 cm absorption (Turner 1993).

As mentioned above, we neglected the hyperfine splitting of the H_2CO transitions in our radiative transfer treatment. This approximation is strictly valid only when the separation of the hyperfine lines is negligible compared to the line broadening. For linear molecules with large splittings, e.g., HCN or N_2H^+, detailed treatments have shown that the total opacity of rotational lines can be uncertain by up to a factor of two (for $\tau_{tot} \gg 1$) if the hyperfine structure is neglected (Daniel et al. 2006). In the case of H_2CO, accurate hyperfine collisional rates are not available because the two-spin recoupling formalism (see Daniel et al. 2004) has not yet been extended to asymmetric tops. However, we assessed the effect of the hyperfine structure by resorting to the approximation that hyperfine rates are proportional to the degeneracy ($2F' + 1$) of the final hyperfine level. Ad hoc selection rules such as $\Delta F = 0$ were also tested at both low and high opacitites ($0 < \tau_{tot} < 50$). Line overlap was included using the absorption probability method (Lockett & Elitzur 1989). The total opacity and excitation temperature (common to all hyperfine levels) were found to agree to within 10% with calculations considering only the rotational structure. As a result, in contrast to molecules containing a ^{14}N nucleus, the hyperfine structure in H_2CO can safely be neglected.

Based on the previous considerations, grids of models were constructed by varying the H_2 density between 10^3 and 10^6 cm^{-3} and the kinetic temperature between 7 and 12 K, in accordance with the known physical structure of B68 (Bergin et al. 2006); the ortho-H_2CO column density was varied between 10^{11} and 10^{15} cm^{-2}, and the OPR of H_2 between 0 and 3, with the objective of reproducing both the total opacity τ_{tot} and the excitation temperature T_{ex} reported in Sect. 2. The line width was fixed at 0.294 km s^{-1}, as deduced from the CLASS HFS fit. It should be noted that the impact of temperature variation (7–12 K) was found to be negligible in our modelling. In the following, we therefore assume a constant temperature of 10 K. On the other hand, the ortho-H_2CO column density was found to be very well constrained by our observations, with a value of $2.2 \pm 0.8 \times 10^{13}$ cm^{-2}. Interestingly, both the excitation temperature and line total opacity were found to depend strongly on the assumed H_2 density and OPR. This is illustrated in Fig. 3, which shows the predicted opacity and excitation temperature as a function of the H_2 density, assuming an OPR equal to zero. In

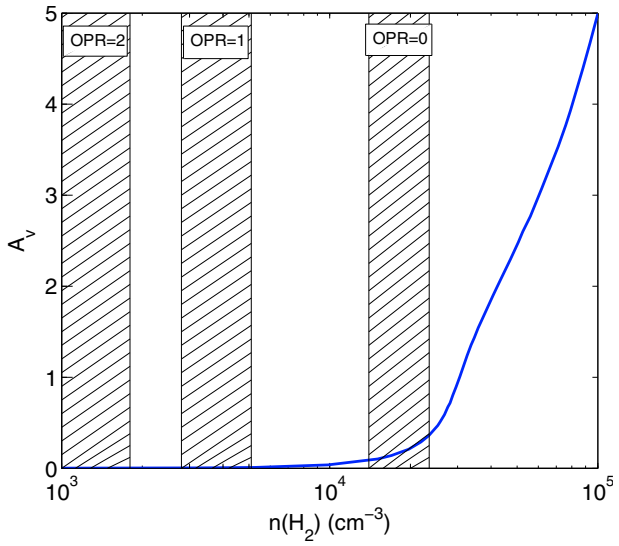

Fig. 4. Visual extinction A_v in B68 as a function of H_2 density adopted from the model of Alves et al. (2001). The dashed areas denote the ranges of H_2 densities compatible with our H_2CO observation for OPR equal to 0, 1, and 2, respectively.

this particular case, our observations constrain the H_2 density to the narrow range 1.4×10^4–2.4×10^4 cm^{-3}.

For a higher OPR, the observed τ_{tot} and T_{ex} can still be reproduced simultaneously but for much lower H_2 densities. This is shown in Fig. 4, where we plot the allowed range of densities for an OPR of 0, 1, and 2 respectively. It should be noted that the dependence of our model on the OPR is linear and not logarithmic. As a result, we can obviously not discriminate between an OPR of zero and, e.g., 10^{-3}. We also report in Fig. 4 the visual extinction of B68 as a function of density, as derived by Alves et al. (2001). The case OPR = 3 is not plotted because it is not compatible with the observations. This figure shows that the H_2 densities compatible with our H_2CO observation relate to the low visual extinction, $A_v \lesssim 0.5$ mag, that is in the outer parts of the core. In the following paragraph, we discuss the implications of this result.

4. Discussion

Our observations and modelling have demonstrated that the H_2CO abnormal absorption originates in regions of the core of moderate extinction. However, the precise origin of the absorption depends on the H_2 OPR. Conversely, one can place constraints on the OPR from simple arguments about the chemistry of the molecule. In particular, it is unlikely that the absorption of H_2CO occurs in regions where $A_v < 0.1$ mag. In these regions, the external UV field dissociates molecules and forms a photon-dominated layer. For example, in their detail modelling of the photon-dominated region (PDR) surface layer of B68, Pineda & Bensch (2007) found that the CO abundance peaks at higher visual extinctions ($A_v \sim 0.4$ mag). Although Pineda & Bensch do not present predictions for H_2CO, we expect its abundance to peak at a similar or slightly greater A_v because, unlike CO, H_2CO should not be self-shielded from the UV field. Deeper inside the cloud, the abundance of both molecules are expected to decrease, because of the freeze-out on grain mantles (e.g., Bergin et al. 2002; Maret & Bergin 2007, in the case of B68). If we assume that the observed H_2CO originates in a region where $A_v \sim 0.4$ mag, our modelling indicates that the OPR is close to 0 (see Fig. 4). As mentioned above, it is very unlikely that the absorption originates in a region where $A_v < 0.1$ mag, and therefore the OPR is necessarily smaller than 1. Thus, our modelling and observations are consistent with $0 \leq \mathrm{OPR} \lesssim 1$, that is with H_2 mostly in para form.

Furthermore, we have shown that the absorption originates in regions where $A_v \lesssim 0.5$ mag, that is the ortho-H_2CO 6 cm absorption line probes the outermost envelope of B68. This conclusion was first reached by Minn & Greenberg (1975). Additional evidence supporting this scenario was subsequently obtained by Henkel et al. (1981) for TMC-1 and by Vanden Bout et al. (1983) for a sample of seven dark clouds. It is interesting to note that our estimate of the ortho-H_2CO column density in B68 ($2.2\pm0.8\times10^{13}$ cm^{-2}) is consistent with the range obtained by the previous authors (10^{12}–10^{14} cm^{-2}). Assuming $A_v = 0.4$ mag and using the relation $N(H_2)/A_v = 0.94\times10^{21}$ cm^{-2} mag^{-1} (Frerking et al. 1982), we obtained an H_2CO relative abundance (in the absorption region) of $X(H_2CO) \sim 6 \times 10^{-8}$. In the envelope of dark clouds, such a large abundance cannot be reproduced by gas phase chemistry models, as first pointed out by Federman & Allen (1991). In the gas phase, H_2CO is mainly formed by the reaction of atomic oxygen with CH_3. However, there is never enough CH_3 in the tenuous envelopes of dark clouds and, in addition, H_2CO is rapidly destroyed by photodissociation and by reactions with C^+ (Federman & Allen 1991). Alternatively, H_2CO may form by successive hydrogenation of CO at the surface of the grains (Tielens & Hagen 1982). This mechanism is expected to be more efficient in the innermost regions of the cores, where the depletion of CO is at its highest. Thus, this scenario would require an efficient mixing mechanism to bring the grains from the interior to the surface, where CO and H_2CO could be photo-evaporated. However, turbulence is observed to be small in B68 with both $C^{18}O$ and ^{13}CO line widths being close to the thermal line width (Lada et al. 2003). Complex radial motions have been observed in this source, but their amplitude is also small (<0.05 km s^{-1}, Maret et al. 2007). As a result, we are currently unable to distinguish between gas phase and grain surface chemistry for the H_2CO formation, which has long been enigmatic in a variety of objects (e.g., Roueff et al. 2006). Future studies will address this important issue further. It will be interesting, in particular, to model other observable H_2CO lines, in particular the 2 mm emission ($2_{11} \rightarrow 1_{11}$) and 2 cm absorption ($2_{11} \leftarrow 2_{12}$). These two lines are, however, understood to originate in higher density regions than the 6 cm line (Vanden Bout et al. 1983). Interferometric observations would also clearly be desirable to probe the spatial distribution of H_2CO absorption in B68. We note that such observations were undertaken by Zhou et al. (1990) in a VLA study of the Bok globule B335.

It is finally interesting to compare the OPR that we derive in B68 to other estimates. Based on observations and modelling of the DCO^+ and HCO^+ emission in this source, Maret & Bergin (2007) inferred an OPR of 0.015, a value that is consistent with the value obtained here. Furthermore, Pagani et al. (2009) used N_2D^+, N_2H^+ and ortho-H_2D^+ observations to derive the OPR in the L183 prestellar core, and found that it is 0.1 in most parts of the cloud. Finally, Maret et al. (2009) recently obtained maps of the H_2 OPR in the shocked gas in NGC 1333 cloud, from which they concluded that the OPR in cold, pre-shock gas is lower than 1. Thus, all these studies are consistent with the result presented here, that is H_2 is mostly in para form in cold gas.

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