H$_2$, H$_3^+$ and the age of molecular clouds and prestellar cores

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Measuring the age of molecular clouds and prestellar cores is a difficult task that has not yet been successfully accomplished although the information is of paramount importance to help in understanding and discriminating between different formation scenarios. Most chemical clocks suffer from unknown initial conditions and are therefore difficult to use. We propose a new approach based on a subset of deuterium chemistry that takes place in the gas phase and for which initial conditions are relatively well known. It relies primarily on the conversion of H$_3^+$ into H$_2$D$^+$ to initiate deuterium enrichment of the molecular gas. This conversion is controlled by the ortho/para ratio of H$_2$ that is thought to be produced with the statistical ratio of 3 and subsequently slowly decays to an almost pure para-H$_2$ phase. This slow decay takes approximately 1 Myr and allows us to set an upper limit on the age of molecular clouds. The deuterium enrichment of the core takes longer to reach equilibrium and allows us to estimate the time necessary to form a dense prestellar core, i.e. the last step before the collapse of the core into a protostar. We find that the observed abundance and distribution of DCO$^+$ and N$_2$D$^+$ argue against quasi-static core formation and favour dynamical formation on time scales of less than 1 Myr. Another consequence is that ortho-H$_2$ remains comparable to para-H$_2$ in abundance outside the dense cores.

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1. Introduction

Star formation is one of the most fundamental problems that astrophysicists have been tackling. Briefly, stars form via a long condensation process, which starts from a warm diffuse material (at approx. 8000 K) that turns first into cold neutral atomic gas (at approx. 100 K, \( n \approx 10^{-10} \) particles cm\(^{-3}\)) and then converts itself into molecular material with densities typically in the range \( 10^2 \rightarrow 10^4 \) particles cm\(^{-3}\) and temperatures in the range 10–20 K, if no local heating source is present. This molecular material is turbulent, and, in some places, dense cores (more than \( 10^4 \) particles cm\(^{-3}\)) appear. Some of these cores probably dissolve again because they are not pressure confined or gravitationally bound, others possibly oscillate and some continue to grow to form prestellar cores (more than \( 10^5 \) particles cm\(^{-3}\) \([1,2]\)). Prestellar cores continue to grow until a collapse starts and a protostar appears. The steps preceding the formation of the protostar are not completely understood and the transition from one phase to the next is difficult to observe. Even basic questions remain. The first one is how do molecular clouds form from the diffuse atomic phase? How long does it take? One puzzle is the formation of H\(_2\) molecules. As long as the medium is not very dense, it takes a very long time for H atoms to meet at the surface of grains, the necessary catalysts \([3]\), and there must be a way to accelerate this process. Indeed, Glover & MacLow \([4]\) suggest that, taking turbulence into account, conversion of H to H\(_2\) should last less than 1–2 Myr. Once the molecular cloud is formed, the details of the first steps of the star formation process remain uncertain. What causes the contraction of the prestellar core and its subsequent collapse? Is contraction due to supersonic turbulence fast decay \([5,6]\) or is it due to quasi-static contraction such as that driven by slow ambipolar diffusion in a magnetically subcritical cloud \([7–9]\) or a combination of turbulence and ambipolar diffusion \([10,11]\)? Which initial or evolving conditions set up the final mass of a star? How do multiple systems form? All these questions were asked long ago and are still present. It is obviously difficult to measure constraining parameters such as the magnetic field strength, the cosmic ray ionization rate and its partly related dark cloud electronic abundance, which would help to assess the importance of ambipolar diffusion (however, see Crutcher \textit{et al.}[12]). We refer the reader to recent reviews \([1,13]\) for more detailed discussions on these topics.

Because the different prestellar core-making theories imply different time scales (by a factor of up to 10–20), measuring the age of the clouds and of the cores within them could help us to discriminate between slow and fast theories. In principle, chemical clocks could be used because different species appear with different time scales, and measuring the abundance ratio of such species would provide the needed information, but many problems remain to be solved—in particular, the initial conditions are usually ill-defined. For example, many attempts to establish a clock based on sulphur chemistry have led to contradictory results. Hatchell \textit{et al.}[14] found that SO and SO\(_2\) disappear early whereas CS reaches maximum abundance at steady state in hot cores, contrary to what was found previously \([15–17]\), the main difference being in the chosen initial conditions. Several of these authors reached the conclusion that it is difficult to build chemical clocks (see also \([18]\)). The existence of chemical clocks is therefore not yet established.
We recently suggested two new possible clock representations to derive the age of clouds and prestellar cores [19–21], i.e. the time that has elapsed since their formation (we arbitrarily fix the birth—or zero age—of the cloud to the moment when all H is converted to H2, though this is probably not a well-defined moment). The first possible clock is linked to the direct detection and measurement of grown grains in dark clouds (the coreshine effect [22]), which we have shown to be feasible in many clouds [20]. These big grains have been built up from interstellar standard grains, and growth models such as those of Ormel et al. [23] allow us to estimate the time necessary for their formation.

The second method involves the chemistry of deuterium. The elemental D/H ratio being approximately $1.5 \times 10^{-5}$ in the vicinity of the solar system [24], it could be expected that this ratio is also found in H-containing molecules. Consequently, the deuterated species should often be undetectable when the main species is already rare. However, deuterated species were discovered early on (DCN in Orion by Jefferts et al. [25]), implying large deuterium enrichments. Even species with double and triple D substitution were detected against all odds, as their abundance should have been $10^{10}$ and $10^{15}$ times lower than that of the main isotopologue. This is the case for ND$_3$ [26–29] and CD$_3$OH [30]. These species are, most of the time, found in depleted cores, i.e. cores where most species such as CO, CS, SO... have stuck to the grains to form ice [31–35]. Deuterated species also appear in depleted cores because these cores are very cold (which is a necessary condition for depletion as well). Therefore, they are not expected elsewhere or in a warm environment except when a newborn star at the centre of the core has just evaporated the ice from nearby grains, which have also been enriched in deuterium (deuterated methanol and formaldehyde are thought to form on grains [36–38]). The deuterated species then are thought to exist out of equilibrium for a short while. A counterexample of ‘warm’ deuterium chemistry towards the Orion Bar is, however, discussed by Parise et al. [39]. Most studies on deuterium chemistry have been carried out based only upon the hypothesis of a cold and depleted medium. H$_2$D$^+$ alone was first considered as a deuteration vector but was necessarily not sufficient to produce triply deuterated species at a high abundance level, and it was Lis et al. [26] who proposed including D$_2$H$^+$ and D$_3^+$ in their chemical model to explain their detection of ND$_3$. Roberts et al. [40] explored in more detail the consequences of this extension, but some deuterated species were overproduced in their models. This is because these models have ignored the (ill-known) state-sensitive chemistry of H$_3^+$, which depends on the ortho-H$_2$ abundance. Although the abundance of ortho-H$_2$ was discussed when the first astrochemical models were developed [41], and discussed by Flower & Watt [42], it was not until 1991 that the importance of the ortho/para spin states of H$_2$ in the deuterium chemistry was actually put forward by Pineau des Forêts et al. [43] but considering only para-H$_2$D$^+$. The role of ortho-H$_2$ in the ortho/para H$_2$D$^+$ ratio was discussed in detail by Pagani et al. [44]. In the same paper, the possibility that the abundance of H$_2$D$^+$ is equal to that of H$_3^+$ was also suggested. Although this hinted at the possibility of detecting very high abundances of deuterated isotopologues, this result, which arrived too early, remained unnoticed. We consider here another consequence of the presence of ortho-H$_2$ for the spatial dependence of observed deuterated species, such as N$_2$D$^+$ and DCO$^+$, in assessing the age of molecular clouds and the time scale of formation of prestellar cores [21].
2. The deuterium chemistry

In molecular clouds, the most efficient path to inject deuterium in the chemical network is to extract it from HD, which is its reservoir, by successive reactions with $\mathrm{H}_3^+$ isotopologues [40],

\[
\begin{align*}
\mathrm{H}_3^+ + \mathrm{HD} &\rightleftharpoons \mathrm{H}_2\mathrm{D}^+ + \mathrm{H}_2 + 232 \text{ K}, \\
\mathrm{H}_2\mathrm{D}^+ + \mathrm{HD} &\rightleftharpoons \mathrm{D}_2\mathrm{H}^+ + \mathrm{H}_2 + 187 \text{ K}
\end{align*}
\]

and

\[
\begin{align*}
\mathrm{D}_2\mathrm{H}^+ + \mathrm{HD} &\rightleftharpoons \mathrm{D}_3^+ + \mathrm{H}_2 + 234 \text{ K}.
\end{align*}
\]

The backward reactions are endothermic with an energy barrier of about 200 K (when considering only the ground level for each species) and were thought to be negligible at the low temperatures found in prestellar cores (less than or equal to 20 K), in which case the abundance ratios

\[
\frac{[\mathrm{H}_n\mathrm{D}_{3-n}^+]}{[\mathrm{H}_{n+1}\mathrm{D}_{2-n}^+]}_{n=0,1,2}
\]

would be greatly enhanced. However, such enhancement can be limited by various processes:

- dissociative recombination of $\mathrm{H}_3^+$ (and its deuterated counterparts) with free electrons or negatively charged grains;
- reactions with ‘proton-friendly’ molecules such as CO and N$_2$ that destroy the trihydrogen cations to produce HCO$^+$ and N$_2$H$^+$; and
- ortho-H$_2$ that can react with the deuterated trihydrogen cation and remove the deuterium.

The role of the ionization fraction (first item) has been discussed in the Watson scheme [45]. The deuterium fractionation of HCO$^+$ was subsequently used to measure electronic abundances in dense cores (an important parameter to estimate the ability of the magnetic field to prevent the core collapse [46]), but the impact of ortho-H$_2$ on the abundance of DCO$^+$ was ignored. Therefore, this scheme is no longer valid today, and electronic abundances cannot be retrieved accurately this way.

The second item indicates that depleted cores are preferable while the last item was ignored for a long time because the ortho-H$_2$ abundance was considered to be negligible in dark clouds. However, ortho-H$_2$ could play an important role: its internal energy (for $J = 1$) is 170 K above the ground level ($J = 0$) of para-H$_2$. Because the ortho-H$_2$D$^+$ ground state ($J_{kk'} = 1_{11}$) is 84 K above the para-H$_2$D$^+$ ground state ($J_{kk'} = 0_{00}$), the reverse reaction (2.1) becomes possible if both the reactants are in their ortho state. Their total internal energy (170 + 84 K) exceeds the endothermicity of the backward reaction, which can therefore occur. Hugo et al. [47] has also indicated that para-H$_2$D$^+$ would react with ortho-H$_2$ to produce ortho-H$_2$D$^+$. Thus, if ortho-H$_2$ is not negligible compared with para-H$_2$, a large amount of para-H$_2$D$^+$ is converted first to ortho-H$_2$D$^+$ and then back to para-H$_3^+$ and the deuteration route is closed. The importance of ortho-H$_2$ on the deuteration efficiency of a typical species, N$_2$H$^+$, has been numerically explored.

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in Pagani et al. ([19]; see also Flower et al. [48]). Several other works address the spin-state chemistry of $H_3^+$ isotopologues [49–51]. However, they focus on the steady-state case where ortho-$H_2$ has dropped enough to play a minor role, which is probably not relevant in most cases.

Once $H_3^+$-deuterated isotopologues become abundant enough, two routes for deuteration open—one in the gas phase, and one on the grains. The first one is simply reactions with $H_2D^+$, etc., such as

\[
H_3^+ + N_2 \rightarrow N_2H^+ + H_2,
\]

\[
H_2D^+ + N_2 \rightarrow N_2D^+ + H_2 \quad \text{(for 1/3),}
\]

\[
\rightarrow N_2H^+ + HD \quad \text{(for 2/3),}
\]

\[
D_2H^+ + N_2 \rightarrow N_2D^+ + HD \quad \text{(for 2/3),}
\]

\[
\rightarrow N_2H^+ + D_2 \quad \text{(for 1/3)}
\]

and

\[
D_3^+ + N_2 \rightarrow N_2D^+ + D_2.
\]

DCO$^+$ and HCO$^+$ are similarly produced with CO instead of $N_2$ in the above reactions. The second route comes from the fact that $D_3^+$ is the terminal species and, as such, it can only be destroyed by reactions with neutrals (CO, $N_2$, ... ) or recombination with electrons. The recombination enriches the cloud in atomic deuterium. Roberts et al. [40] show that the $D/H$ atomic ratio can reach 0.3, which is the required ratio to produce triply deuterated methanol onto grains.

3. The age of molecular clouds

Although deuterated species are seen mostly in depleted cores, the condition that CO is rare enough to let the deuterium chemistry develop is partly wrong. Indeed, if CO is undepleted, its typical abundance in dark clouds is $1–2 \times 10^{-4}$ while that of HD is approximately $3 \times 10^{-5}$ [24]. In which case, $H_3^+$ would react five times more often with CO than with HD, somewhat quenching the production of $H_2D^+$. However, a little production of $H_2D^+$ still takes place, but it will again react preferentially with CO. It is easy to see that the production of DCO$^+$ is in first approximation independent of the abundance of CO and $H_2D^+$, as their abundances are inversely proportional to each other’s (if CO abundance decreases, $H_3^+$ will react more often with HD and $H_2D^+$ abundance will increase). Therefore, DCO$^+$ should be equally abundant everywhere in clouds. This is not the case for $N_2D^+$, as the reaction probability of $N_2$ with $H_3^+$ isotopologues is strongly reduced in a CO-rich environment. Moreover, $N_2D^+$ itself is destroyed by CO when CO is not depleted. In fact, we can show for a chemical model in steady state that DCO$^+$ should be most abundant for CO abundances in the range $1 \times 10^{-5}–2 \times 10^{-4}$ [21] and not where CO is strongly depleted. It is clearly not so; DCO$^+$ does not seem to be observed outside depleted cores (see discussion in Pagani et al. [21]). The only explanation we can find is that ortho-$H_2$ is still abundant in the cloud. The question therefore becomes how old can the cloud be before ortho-$H_2$ drops to a level low enough to allow deuteration to proceed?
There exist two parallel processes that convert ortho-H$_2$ into para-H$_2$
[41,42,48,52],
\[
o-H_2 + H^+ \rightarrow p-H_2 + H^+ + 170 \text{K} \tag{3.1}
\]
and
\[
o-H_2 + H_3^+ \rightarrow p-H_2 + H_3^+ + 170 \text{K}. \tag{3.2}
\]

In low-temperature clouds, these reactions are only going forward. Reaction (3.1) has been recently revisited by us, and a new rate coefficient has been accurately calculated using a time-independent quantum mechanical approach [53,54]. However, an error has just been discovered, and the new rate, instead of being a factor of approx. 2 higher than the previous one [55], is in fact a factor of approximately 2 lower [56]. The results presented here take into account the corrected rate (a factor of 3.571 lower with respect to the value in Honvault et al. [53,54]). Reaction (3.2) has been studied only in a statistical way [47,57]. Recent experiments have been conducted in the laboratory but are not yet applicable to our very-low-temperature case [58,59]. The two reactions have both low rates, a factor of approximately 5 (for H$_3^+$, on average, approx. 3 for p-H$_3^+$ and approx. 10 for o-H$_3^+$) and a factor of approximately 20 (for H$^+$) below Langevin’s rates. The dominant reaction depends on the relative abundances of H$^+$ and H$_3^+$, which are also relatively close to each other. As ortho-H$_2$ is initially the most abundant species and as both H$^+$ and H$_3^+$ are relatively rare, it takes over a million years for ortho-H$_2$ to decrease to parity with para-H$_2$ and about 5 million years to drop to 10 per cent. Because this is comparable to the expected cloud lifetime, this is a useful clock if one can consider that most of the molecular hydrogen formed in a short period at the birth of the molecular cloud is still an open question [3,4].

Finally, we find that, after 7.5 million years (instead of 6 Myr in Pagani et al. [21]), ortho-H$_2$ has become so rare that DCO$^+$ should be abundant enough to be detectable at any place in dark clouds (figure 1). This age will decrease for cosmic ray ionization rates higher than $1 \times 10^{-17} \text{s}^{-1}$ (figure 2) or if the ortho/para H$_2$ ratio is initially lower than 3 (figure 1). Because the time to form H$_2$ could be long enough to let ortho-H$_2$ disappear on the same time scale, we also ran the model starting from fully atomic H (figure 2). The result is unchanged for DCO$^+$ detectability. No deep integrations of DCO$^+$ outside depleted cores have been performed yet to compare with our predictions. We have pending observations with the GBT.

4. The age of prestellar cores

There are several competing theories to explain the formation of prestellar cores [13,60–62]. In the main stream, two theoretical pictures have emerged: in the first quasi-static picture, cores are thought to be initially supported against their self-gravity by magnetic or turbulent pressure, and progressively evolve towards higher degrees of condensation through either ambipolar diffusion [7,63] and/or the dissipation of turbulence [64]. The requested time for such quasi-static contractions reaches up to approximately 10 Myr, though most of the time is spent before the core becomes observable, the core stage before gravitational collapse being itself limited to approximately 1 Myr [11]. In the
Figure 1. Variation of DCO$^+$ and ortho-H$_2$ abundances in a pseudo-time-dependent chemical model for different starting H$_2$ ortho/para ratios (OPRs) from $3 \times 10^{-3}$ up to 3 and from full atomic initial conditions. The abundance of the sum of the deuterated H$_3^+$ isotopologues is also traced as a thick grey line (for the case H$_2$ OPR = 3). The horizontal dotted line in the lower box indicates the limit of detection of DCO$^+$, and the circles in the upper box mark the corresponding OPR values. Adapted from Pagani et al. [21] with the rate of reaction (3.1) corrected.

second picture, cores form dynamically through local density enhancements owing to supersonic turbulent flows, which can eventually collapse if they are large enough [5,6,62,65]. They result in a fast contraction (less than 1 Myr), close to the free-fall time. Determining the age of prestellar cores would therefore be valuable to help select the correct models. In order to progress in the understanding of this phase, two complementary approaches are currently being developed: comparative/statistical studies of many prestellar cores or detailed studies of individual ones. The lifetime of prestellar cores can be estimated via the statistical studies [66–68], but this has not proved to be very constraining. Measuring the actual age of individual cores requires a detailed modelling of their
properties. Dynamical models suffer from a lack of information (in particular on the magnetic field shape and strength), and chemical models depend very much on initial conditions, not forcibly known. Deuterium chemistry, which is limited to prestellar core regions (possibly containing a newborn protostar), stands apart: (i) inside the prestellar core, depletion limits the number of intervening species; (ii) the \( \text{H}_2 \) ortho/para ratio is very likely to be equal to the statistical ratio (i.e. 3) at formation on the grains; and (iii) the deuteration tracer pair we use, the diazenylium isotopologues \( \text{N}_2\text{H}^+ \) and \( \text{N}_2\text{D}^+ \), appears only inside the prestellar core, being efficiently destroyed by \( \text{CO} \) outside [1]. This has the advantage that these species are not polluted by extended emission (unlike, for example, \( \text{HCO}^+ \)) and have a zero abundance (at least not detectable) at the start of the core contraction, as \( \text{CO} \) depletion has not taken place yet. There remain two input parameters that are not accurately known—the cosmic ray ionization rate and the grain size distribution, which both act on the ionization degree of the cloud and on the overall chemical evolution rate. The latter is now a tractable problem, thanks to the discovery of coreshine, while the former might be estimated following works such as Caselli et al. [69] and more recently Padovani et al. [70], Gibb et al. [71] and Indriolo & McCaIl [72]. Somewhat related is the work by Fontani et al. [73], which uses the \( \text{N}_2\text{D}^+/\text{N}_2\text{H}^+ \) ratio to evaluate the evolution of massive prestellar objects.

The \( \text{N}_2\text{D}^+/\text{N}_2\text{H}^+ \) ratio profile varies along the dense core radius, for example in the well-studied source L183 [74]. Higher density shortens the chemical time scales. If the dense core had remained in the present situation with the present density profile since the start of the deuteration process, the inner part of the core would reach higher deuteration enrichment than the outskirts, as is observed. However, assuming deuteration has started everywhere at the same time, it would reach the observed ratio much earlier in the inner part of the core than in the outskirts. In other words, it would result in a relative central to edge

Figure 2. Variation of \( \text{DCO}^+ \) abundance in a pseudo-time-dependent chemical model for different cosmic ray ionization rates from \( 1 \times 10^{-18} \) up to \( 1 \times 10^{-16} \). The horizontal dotted line indicates the limit of detection of \( \text{DCO}^+ \); the arrows mark the time at which \( \text{DCO}^+ \) abundance crosses the detection limit. Adapted from Pagani et al. [21] with the rate of reaction (3.1) corrected.
deuterium enrichment much higher than observed. A much better match to the observations is obtained if the core has evolved to the present state from a medium originally uniform in density. In the latter case, the densest parts of the core have reached their present density only recently, and the acceleration of the chemistry is just enough to provide the observed larger deuterium enrichment in the centre. Therefore, the static density model gives only a lower limit to the age of the prestellar core. In L183, this lower limit is \(2 \times 10^5\) years, which is compatible with all dynamical models. Again, the chemical models can help us to discriminate between slow and fast contraction cases because the time for ortho-H\(_2\) to disappear is comparable to the dynamical ages we want to measure. For the standard grain size (0.1\(\mu\)m radius on average) and standard cosmic ray ionization rate (\(\zeta = 1 \times 10^{-17}\) s\(^{-1}\)), the time to reach steady state (and obtain an almost density-independent deuteration ratio across the core) is several million years, enough to differentiate between models. To go further, we have coupled our chemical model to our dynamical model [75]. Preliminary results (figure 3) show that only fast contracting models can provide profiles close enough to our observations, while slow models provide too much deuteration. The models shown here reached the observed density profile in approximately 0.5 and approximately 5 Myr, respectively, and therefore the core formed in only approximately 0.5 Myr. These calculations start with a H\(_2\) ortho/para ratio of 3, which would imply that the core started contraction simultaneously with the formation of the cloud. This gives us a lower limit on the age of the cloud of approximately 0.5 Myr. We can also put an upper limit to the age of this cloud: if the contraction starts several million years after the formation of the cloud, we know that ortho-H\(_2\) will

Figure 3. Comparison between fast and slow models, output for the peak density matching the observations, and the \(N_2D^+/N_2H^+\) abundance ratio derived from the observations. The error bars represent the maximum (resp. minimum) ratios using the maximum (resp. minimum) \(N_2D^+\) abundance profile compared with the minimum (resp. maximum) \(N_2H^+\) abundance profile as displayed in fig. 4b of Pagani et al. [74]. The calculations are preliminary (pending corrections), but the situation is clear-cut in favour of the fast model.
have already substantially diminished; in which case, even in the fast contracting model case, the deuteration will go too far (not shown here). The upper limit for the start of the core contraction is probably about 1 Myr, while the ortho/para ratio of H$_2$ remains around 1. This would give us an upper limit (1.5 Myr) for the age of the cloud since its formation. Increasing the cosmic ray ionization rate would accelerate the chemistry and slow down the contraction because of a stronger heating of the cloud, making it more difficult to compress. We would therefore get again too much deuteration. Our chosen value of $\zeta = 1 \times 10^{-17}$ s$^{-1}$ is already at the lower range of possible values [70]. An even lower $\zeta$ value seems difficult to advocate. Therefore, slowing down the chemistry to allow time to slow contracting models to build up the observed core without overproducing N$_2$D$^+$ does not seem possible.

5. Conclusions

The long decaying ortho/para H$_2$ ratio and its influence on deuterium enrichment make gas-phase deuterium chemistry a useful tool to measure the age of clouds and prestellar cores. Absence of widespread DCO$^+$ in cloud envelopes and low abundance of N$_2$D$^+$ in the outer layers of dense cores both indicate that the clouds are young and cores cannot be formed via quasi-static contraction. As a consequence, ortho-H$_2$ should still be abundant in the low-density parts of the clouds. To better assess these results, progress should be made on the paramount state-conversion reaction of H$_2$ with H$_3^+$, and the cosmic ray ionization rate should be constrained with precision.

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