Hyperfine-resolved transition rates of the DCO\(^+\) ion by collisions with helium

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ABSTRACT

The importance of rotational transition rates in the analysis of cold interstellar clouds is well known. We present results, for temperatures ranging from 5 to 80 K, for the hyperfine-resolved rotational transitions of DCO\(^+\) induced by collision with helium. Since the isotopic substitution is not expected to introduce significant changes, close-coupling calculations are based on a potential energy surface obtained for He–HCO\(^+\) and checked by accurate pressure broadening and shift measurements. The well-grounded assumption that deuterium nuclear spin is not affected by the collisions allowed us to obtain the hyperfine-resolved transition matrix elements as a sum of spin-free transition matrix elements, which account for collision dynamics, multiplied by purely geometrical factors, which account for the hyperfine dependence. The temperature dependence of the rates is weak for downward transitions \(j \rightarrow j', j < j\), while for upward transitions \(j' > j\) it can be large due to the need of energy to be transferred from translation to rotation. The dependence of the rates on \(j\) and \(j'\) and hyperfine propensities is discussed. The rates for quasi-elastic purely hyperfine transitions \(j, F \rightarrow jF'\) are also obtained.

Key words: molecular data – scattering – ISM: molecules.

1 INTRODUCTION

The rates of rotational excitation and de-excitation of molecules and molecular ions are crucial in modelling molecular clouds. Without these rates, only approximate estimates of the column densities are possible assuming local thermodynamic equilibrium (LTE), which at the typical densities of dense interstellar clouds is generally not a good approximation (Lique et al. 2009). When rates are known, non-LTE calculations may be performed allowing not only accurate column density estimates, but also estimates of kinetic temperature and volume density. Therefore, astrophysical models improve significantly in sophistication with the knowledge of collision rates.

While, in practice, due to the larger abundance of molecular hydrogen, collisions with para-H\(_2\)(\(j = 0\)) are more likely in molecular clouds compared to collisions with He atoms, most part of rate calculation were performed for collision with helium. Indeed, calculations with helium are simpler and are commonly used to model collisional excitation coefficients with H\(_2\) by applying an appropriate scaling factor. However, this is an approximate method: ratios between 2 and 4 were found by Monteiro (1985) for the case of HCO\(^+\). Due to the astrophysical interest of HCO\(^+\), close-coupling calculations for the state-to-state rotational transition rates of this ion by collisions with helium were recently published by Buffa, Dore & Meuwly (2009, and references therein). They were based on an accurate potential energy surface (PES) validated by an experimental and theoretical study of pressure broadening and shift of HCO\(^+\) rotational lines perturbed by collisions with He (Buffa et al. 2008).

The deuterated isotope DCO\(^+\) has also astrophysical relevance. It was observed in interstellar clouds (Wootten, Loren & Snell 1982; Butner, Lada & Loren 1995; Pfalzner 2004; Caselli & Dore 2005; van der Tak et al. 2009) and in circumstellar discs (van Dishoeck, Thi & van Zadelhoff 2003). Moreover, DCO\(^+\) has been surveyed in a large sample of starless core by Lee, Myers & Plume (2004) with the aim of detecting infall motions.

Therefore, we extend here to this isotope the study of state-to-state transition rates. DCO\(^+\) is characterized by the hyperfine structure induced by deuterium nuclear spin \(I = 1\). Indeed, even though hyperfine energy is far smaller than rotation energy, hyperfine effects must be taken into account when dealing with collisions. The problem was first raised by the observation of anomalous intensities of hyperfine components in the HCN (Monteiro & Stutzki 1986, and references therein), NH\(_3\) (Stutzki & Winnewisser 1985a, b) and HC\(_3\)N (Churchwell, Walmsley & Winnewisser 1977; Tolle et al. 1981) spectra coming from interstellar clouds, which could be explained only when the hyperfine dependence of collisional rates was taken into account. Hyperfine-resolved rates were recently studied for CN (Lique & Klos 2011), HCl (Neufeld & Green 1994), NO (Lique et al. 2009) and HCN/HNC (Faure et al. 2007). Close-coupling calculations for \(N_2H^+\)--He hyperfine-resolved transition rates allowed extension of this kind of study to diazenylum which...

Recently, van der Tak et al. (2009) have shown that hyperfine effects are especially interesting for deuterium-bearing molecules, such as DCO⁺, because they are good probes of the physical and chemical structure of molecular cloud cores on the verge of star formation.

2 THEORY

In order to calculate the hyperfine-resolved transition rates, we use the recoupling technique by Corey & McCourt (1983) and Corey (1984). It assumes that hyperfine energy is negligible with respect to rotational energy and that nuclear spin I is unaffected by collisions. Hence, any collisional change of the hyperfine quantum number \( F = I + J \) is due only to changes of the amplitude or of the orientation of rotational momentum \( J \). As a consequence, the hyperfine-resolved transition matrix elements are obtained by a sum of spin-free transition matrix elements, which account for collision dynamics, multiplied by purely geometrical factors, which account for the hyperfine dependence.

We denote the spin-free transition matrix elements by \( T_{R,l',J'}^{K,F}(E) \), where \( l \) is orbital angular momentum,

\[
K = l + J = l' + J'
\]

is total angular momentum and \( E = E_r + E_i \) is the energy of the colliding pair. \( E_r \) is the ingoing rotational energy of the ion and \( E_i \) is the ingoing translation energy of orbitonal motion. The cross-section for the \( j \rightarrow j' \) transition is given by

\[
\sigma_{j,j'}(E_i) = \frac{\pi}{k^2} \sum_k \frac{2K + 1}{2J + 1} \sum_{l'l'} |T_{j,j'}^{K,F}(E)|^2,
\]

where \( k = (2\mu E_r)^{1/2} \) is the wavevector and \( \mu \) is the reduced mass of the colliding pair.

Recoupling allows us to obtain the \( F \)-resolved transition matrix elements:

\[
T_{F,F'}^{l,l'}(E) = (-1)^{l-l'+F-F'}[(2F + 1)(2F' + 1)]^{1/2} \sum_{K} \frac{2K + 1}{2J + 1} \sum_{l'l'} |T_{j,j'}^{K,F}(E)|^2,
\]

where \( \{a,b,c\} \) are 6-j coefficients and \( J = I + F = I' + F' = K + I \) is the total angular momentum, including nuclear spin. The hyperfine-resolved cross-sections are

\[
\sigma_{J,F,F'}(E_i) = \frac{\pi}{k^2} \sum_J \frac{2I + 1}{2F + 1} \sum_{l'l'} |T_{J,F,F'}^{l,l'}(E)|^2.
\]

By equations (3) and (5), one can see that hyperfine-selective cross-sections, at difference with spinless cross-sections, contain not only the absolute squares of rotational transitions matrix elements \( |T_{j,j'}^{K,F}(E)|^2 \), but also interference between different \( K \) values.

3 SPIN-FREE RESULTS

The scattering matrix elements \( S_{J,F,F'}^{K,F}(E) \) were calculated within the close-coupling theoretical framework proposed by Arthurs & Dalgarno (1960) and implemented in the MOBCAT code (Hutson & Green 1994).

![Figure 1. Jacobi coordinates for HCO⁺–He and DCO⁺–He potentials.](https://academic.oup.com/mnras/article-abstract/421/1/719/990580/fig1)
From Fig. 2, one can see that the temperature dependence is not large for the downward rates: changing $T$ by a factor of 16, one has a change of $q$ that in some cases can be by a factor of 2, but in most cases is only a few per cent. When $T$ increases, the increase of the collision velocity $\bar{v}$ is balanced by the decrease of the cross-section $\sigma$. Different is the case of upward rates: due to the Boltzmann factor in equation (9), they strongly decrease when the energy jump involved in the transition is large with respect to $k_B T$.

From Fig. 2, one can also see that the rates depend more on $\Delta j = j' - j$ than on $j'$. This means that the downward rates depend more on the change of angular momentum than on the change of energy. For instance, the rates of $\Delta j = -1$ transitions (black lines) do not change much with $j'$ even though the energy involved is proportional to $j' + 1$. Moreover, the transition $j = 3 \rightarrow 0$ (green line in Fig. 2a) involves approximately the same energy jump ($\sim 14.42$ cm$^{-1}$) as the transition $j = 6 \rightarrow 5$ (black line in Fig. 2f), but the $3 \rightarrow 0$ rate is about six times smaller than the $6 \rightarrow 5$ rate. The decrease of $q_{j,j'}$ for increasing (absolute) values of $\Delta j$ is well evident. For low $\Delta j$ values ($\Delta j = -1, -2, -3$ and $-4$; black, red, green and blue lines, respectively), such a trend is quite regular, while for higher $\Delta j$ values a preference is found for odd $\Delta j$ rather than for even $\Delta j$. By looking at Figs 2(b)–(e), one can see that the rates are larger for the $\Delta j = -5$ transitions (cyan) than for the $\Delta j = -4$ transitions (blue). The same parity effect is found by comparing in Figs 2(b) and (c) the rates for the $\Delta j = -6$ (magenta) and $-7$ (orange) transitions.

When $\Delta j \leq -4$, the preference for odd parity transitions looks as a systematic trend, as is confirmed by comparing the cases $\Delta j = -8$ (grey) and $-9$ (violet), a comparison which is not evident from Fig. 2(a) due to the small magnitude of both quantities, but which can be done by looking at the Supporting Information in the electronic version of the paper.

Such parity preference is not easily explained by looking at the features of the interaction potential. Indeed, a check changing the magnitude of the radial strength functions $V_\lambda (R)$ for even or odd $\lambda$ values did not yield clear effects for the $\Delta j$ parity preferences of the transition rates.

As far as comparison to the rates of the hydrogenated isotope HCO$^+$ (Buffa et al. 2009, and references therein) is concerned, we could verify that for the downward rates the difference is not large. In most cases, it is a few per cent and rarely it is as large as 20 per cent. It can be larger for upward transitions: at low temperatures, the HCO$^+$ rates are lower than the DCO$^+$ ones because the $\sim 24$ per cent difference of rotational constant $B$ changes the energy jump and the Boltzmann factors in equation (9).

4 HYPERFINE PROPENSITIES

The hyperfine-resolved cross-sections $\sigma_{jF,j'F'}$ were calculated by equation (5) and the rates were obtained by thermal integration:

$$q_{jF,j'F}(T) = \frac{\bar{v}}{(k_B T)^2} \int_0^\infty E_i e^{-E_i/k_B T} \sigma_{jF,j'F}(E_i) dE_i. \quad (10)$$
The hyperfine propensity $p_{jF,j'F'}$ is the ratio between hyperfine-resolved and spinless rates:

$$ p_{jF,j'F'} = \frac{q_{jF,j'F'}}{q_{j'}}, \quad (11) $$

Obviously, one must have

$$ \sum_{F'} p_{jF,j'F'} = 1. \quad (12) $$

Since the $j=0$ state is not split by hyperfine interaction, propensities for transitions from or to that rotational state are trivial:

$$ p_{jF,01} = 1 \quad p_{01;j'F'} = \frac{2F'+1}{3(2j'+1)}. \quad (13) $$

The complete set of numerical values of hyperfine propensities $p_{jF,j'F'}$ for all the downward rotational transitions $1 \leq j < j' \leq 9$ is reported, for $T$ ranging from 5 to 80 K, in Supporting Information in the electronic version of the paper. The propensities for upward transitions can be obtained by

$$ p_{j'F,jF'} = p_{jF,j'F'} \left( \frac{2F'+1}{2F'+1} \right) \frac{(2j'+1)j'}{j+1}. \quad (14) $$

It is well known that the hyperfine propensities depend on the difference $\Delta$ between the change of $F$ and the change of $j$:

$$ \Delta = |\Delta F - \Delta j|. \quad (15) $$

Such a feature is shown in Fig. 3 for the case $j=7 \to 6$. The reduced hyperfine propensity

$$ \hat{p}_{jF,j'F'} = p_{jF,j'F'}/(2F'+1) \quad (16) $$

is plotted for the nine possible hyperfine transitions. The denominator $(2F'+1)$ was introduced in the definition of $\hat{p}$ in order to eliminate degeneracy effect. One can see that the propensity for the three $\Delta = 0$ transitions (solid) is larger by a factor of $\sim 5$ than that for the four $\Delta = 1$ transitions (dash). Moreover, the propensity for the two $\Delta = 2$ transitions (dot–dashed) is still lower by approximately the same factor.

In order to describe how effective is the $\Delta$ dependence of propensities, it is useful to introduce the average reduced propensities $\bar{p}^0$, $\bar{p}^1$, and $\bar{p}^2$, for the three cases $\Delta = 0$, 1 and 2, respectively:

$$ \bar{p}^0_{jF} = \frac{1}{3} \hat{p}_{jF+jF'+1} + \hat{p}_{jF'} + \hat{p}_{jF-1F'-1'}, \quad (17) $$

$$ \bar{p}^1_{jF} = \frac{1}{4} \hat{p}_{jF+jF'+1} + \hat{p}_{jF'} + \hat{p}_{jF-1F'-1'} + \hat{p}_{jF-1F'+1}. $$

$$ \bar{p}^2_{jF} = \frac{1}{2} \hat{p}_{jF+jF'+1} + \hat{p}_{jF-1F'+1}. $$

Fig. 4 performs this kind of analysis for $j=2, 4$ and 6. Solid lines plot $\bar{p}^1_{jF}/\bar{p}^0_{jF}$, (average reduced propensity of $\Delta = 1$ transitions)/(average reduced propensity of $\Delta = 0$ transitions). Dashed lines plot $\bar{p}^2_{jF}/\bar{p}^0_{jF}$, (average reduced propensity of $\Delta = 2$ transitions)/(average reduced propensity of $\Delta = 0$ transitions). A smaller value of these two ratios means more restrictive propensity rules. The case $j=2$ is reported in Fig. 4(a). One can see that, depending on temperature and on $\Delta j$, $\bar{p}^1_{jF}/\bar{p}^0_{jF}$ ranges between 0.3 and 0.7, while $\bar{p}^2_{jF}/\bar{p}^0_{jF}$ ranges between 0.18 and 0.7.

**Figure 3.** Reduced hyperfine propensities for the $j=7 \to 6$ rotational transition. The propensity is larger for the $\Delta j = \Delta F$ transitions (solid) than for the $|\Delta j - \Delta F| = 1$ transitions (dash). It is still smaller for the $|\Delta j - \Delta F| = 2$ transitions (dot–dashed).

**Figure 4.** Solid lines plot the ratio $\bar{p}^1_{jF}/\bar{p}^0_{jF}$, (average reduced propensity of $|\Delta j - \Delta F| = 1$ transitions)/(average reduced propensity of $\Delta j = \Delta F$ transitions). Dashed lines plot the ratio $\bar{p}^2_{jF}/\bar{p}^0_{jF}$, (average reduced propensity of $|\Delta j - \Delta F| = 2$ transitions)/(average reduced propensity of $\Delta j = \Delta F$ transitions). See equation (17).
The case $j = 6$ is reported in Fig. 4(c). One can see that the preference for low $\Delta$ values is much more effective than that found for $j = 2$. $\mathbf{p}_h(j_fj'/j'_f)_{\Delta'j} = 0.14$ and 0.21, while $\mathbf{p}_h(j_fj'/j'_f)_{\Delta'j} = 0.02$ and 0.04.

Fig. 4(b) shows that the trend for the $j = 4$ rotational transitions is intermediate between that found for the cases $j = 2$ and 6.

On the whole, from Fig. 4 one can conclude that preference for $\Delta F = \Delta j$ hyperfine transitions has a rapid increase for increasing values of $j$, while the dependence on $\Delta j$ is by far less marked and less clear. A feature common to the cases (a), (b) and (c), and to other $j$ values not reported in Fig. 4, is that the effectiveness of $\Delta F$ rules is minimum when $\Delta j = -3$ (green).

5 OPACITY TENSOR ANALYSIS

The propensity rules for hyperfine transitions are better understood by resorting to the so-called opacity tensor analysis. This was introduced by Alexanders & Dagdigian (1985) working with the irreducible components of the $\mathbf{T}$-matrix (Orlikowsky & Alexanders 1985), characterized by given values of $j$, the angular momentum transferred by the collision from orbital motion to ion rotation:

$$ j_t = j' - j = l - l'. $$

The $j_t$ decomposition yields

$$ \sigma_{j;j'} = \sum_h \sigma_j^{h} = \sum_h \frac{\pi}{k^2(2 j + 1)} \mathbf{p}_h^{j,j'}, $$

$$ \mathbf{p}_h^{j,j'} = \frac{1}{2 j + 1} \sum_{j'} | T_h^{j;j'} |^2, $$

$$ T_h^{j;j'} = (2 j + 1) \sum_k (2 K + 1)(-1)^{j-l+K} $$

$$ \times \left\{ j \ j' \ j_t \right\} T_k^{j;j'} $$

for the spin-free cross-sections and

$$ \sigma_{j;j'} = \sum_h \frac{\pi(2 F' + 1)}{k^2} \left\{ j \ j' \ j_t \right\} T_h^{j;j'} $$

for the hyperfine-resolved cross-sections. As a consequence of the triangular condition, the sum in equations (19) and (22) extends to $|j - f| \leq j_t \leq j + f$.

Resorting to the $j_t$ decomposition, Alexanders & Dagdigian (1985) could prove the preference for $\Delta F = \Delta j$ transition by supposing that

(i) $j$ and $j'$ are large with respect to $l$,

(ii) $\sigma_j^{h;j'}$ decreases for increasing values of $j_t$.

The relevance of the first condition is well clear from Fig. 4: the effectiveness of $\Delta F$ preferences increases rapidly with $j$.

The relevance of the second condition becomes clear by comparing Figs 4 and 5, which plots versus translation energy $E_t$, the relative contribution to $\sigma_j^{h;j'}$ of $\sigma_j^{h;j'}$ corresponding to the lowest $j_t$ value, $j_t = |j - f|$. In the comparison, it is useful to remember that $T = 80$ K corresponds to an average translation energy $E_t \simeq 70.8$ cm$^{-1}$. One can see that, at fixed $j$, the dependence on $j$ of the effectiveness of $\Delta F$ rules is well explained by the trend of $\sigma_j^{h;j'} / \sigma_j^{h;j'}$. For instance, the minimum effectiveness is found when $\Delta j = -3$ (green lines in both figures) corresponding to a minimum contribution of $\sigma_j^{h;j'}$ to the total cross-section $\sigma_j^{h;j'}$. The temperature dependence observed in Fig. 4 is also well explained by the energy dependence shown in Fig. 5. For instance, the slope of the black lines (e) in Fig. 4 is connected to the slope of the black lines in Fig. 5.

As far as the energy dependence of the cross-sections is concerned, Daniel et al. (2004) observed that for $\text{N}_2\text{H}^+ - \text{He}$ collisions the decrease of $\sigma_j^{h;j'}$ with $j_t$ does not extend at low temperatures for the $j = 2 \rightarrow 5$ transition. Fig. 6(a) shows what we find for the ion we are studying and for the transition $j = 5 \rightarrow 2$. One can see that the contribution of the minimum $j_t$ value (black) is comparable to that of higher $j_t$ values up to $E_t \simeq 100$ cm$^{-1}$, while it becomes dominant at high energies. The decrease of $\sigma_j^{h;j'}$ for increasing $j_t$ values is questionable since the second contribution comes from the maximum $j_t$ value, $j_t = j + f = 7$ (cyan). One can also see that, as observed by Daniel et al. (2004), the contribution of $j_t$ values of the same parity as $\Delta j$ is larger than the contribution of $j_t$ values of opposite parity. The contribution of $j_t = 3, 5$ and 7 (black, green and cyan, respectively) is larger than the contribution of $j_t = 4$ and 6 (red and blue, respectively).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{image.png}
\caption{Relative contribution to $\sigma_j^{h;j'}$ of the lowest $j_t$ value. The ratio $\sigma_j^{h;j'} / \sigma_j^{h;j'}$ is plotted versus translation energy $E_t$.}
\end{figure}
Figure 6. Contribution to the cross-section \( \sigma_{j'j} \) coming from different values of angular momentum \( j \) transferred from orbital motion to ion rotation. Cases \( j = 5 \rightarrow 2 \) (a) and \( 7 \rightarrow 5 \) (b) are shown. In case (b), the contribution of \( j_t = 11 \) and 12 is negligible.

The overall features of the \( j_t \) decomposition depend on \( j \) and \( j' \) and can be different from what we found for the case \( j = 5 \rightarrow 2 \). An instance is shown in Fig. 6(b). For the transition \( j = 7 \rightarrow 5 \), the contribution of the minimum value (\( j_t = |j - j'| = 2 \), black) predominates at all energy values, while the decrease of \( \sigma_{j_t} \) for increasing \( j_t \) values is quite well respected.

6 QUASI-ELASTIC TRANSITIONS

The rates \( q_{jFj'F'} \) for quasi-elastic, purely hyperfine, transitions are reported in Fig. 7 for \( j \) values ranging from 1 to 9. For each \( j \) value, six different hyperfine transitions are possible. Only three of them are displayed because the other are different only by degeneracy factors:

\[
q_{jF,j'F'} = \frac{q_{jF,j'F'}}{2F' + 1}.
\]

The complete set of numerical values of \( q_{jF,j'F'} \) can be found as Supporting Information in the electronic version of the paper.

From Fig. 7, one can see that for low \( j \) values the rates are of the same order of magnitude (\( 10^{-10} \) cm\(^3\) s\(^{-1}\)) as the \( j \) changing transitions reported in Fig. 2, while they become smaller for higher \( j \) values. The discussion in the previous section explains this trend.

Indeed, since \( \Delta j = 0 \) and \( \Delta F \neq 0 \), the rates must tend to zero when \( j \) tends to infinity. In the same way, it is not surprising that for large \( j \) values the rates for \( \Delta F = -2 \) transitions (dash lines) are smaller than the ones for \( \Delta F = -1 \) (solid lines) and 1 (dot–dashed lines) transitions.

7 CONCLUSIONS

We extended to the deuterated species the study performed by Buffa et al. (2009, and references therein) for state-to-state transition rates of HCO\(^+\) induced by collision with helium.

Hyperfine splitting effects were treated by the recoupling technique (Corey & McCourt 1983), which assumes that nuclear spin is unaffected by collisions. This allows us to obtain the hyperfine-resolved transition matrix elements as a sum of spin-free transition matrix elements, which account for collision dynamics, multiplied by purely geometrical factors, which account for the hyperfine dependence. The spin-free scattering matrix was obtained by accurate close-coupling calculations (Hutson & Green 1994) and by a tested interaction potential (Buffa et al. 2008).

The spin-free transition rates \( q_{jj'}(T) \) and the hyperfine-resolved transition rates \( q_{jFj'F'}(T) \) were calculated for \( j \) and \( j' \) ranging from 0 to 9 and for temperatures ranging from 5 to 80 K.
As far as the quasi-elastic \((j = j')\) or downward \((j > j')\) rates are concerned, the trend is dominated by the geometry of angular momenta, while the amount of energy \(\Delta E = E_{j'} - E_j < 0\) transferred from orbital motion to ion rotation plays only a secondary role. Obviously, the situation can be different for the upward rates \((j < j')\) as a consequence of the Boltzmann factor \(e^{-\Delta E/\kappa T}\) in equation (9).

Indeed, for quasi-elastic and downward rates, we found a weak dependence on temperature and \(\Delta E\). Moreover, the difference between the spin-free downward rates of DCO\(^+\) and HCO\(^+\) is small in spite of a ±24 per cent difference of the rotational constant \(B\) of the two isotopes. On the contrary, a strong dependence on \(\Delta j\) and \(\Delta F\) was found. The main features of such a dependence are not surprising, while the origin of some minor details is not well clear.

For the spin-free downward rates, the decrease for increasing values of \(|\Delta j|\) is not surprising, while a less clear minor feature is that when \(|\Delta j| \geq 4\) there is a preference for parity changing transitions (odd \(\Delta j\) values) with respect to parity conserving transitions (even \(\Delta j\) values).

For hyperfine-resolved transition rates, the preference for \(\Delta F = \Delta j\) transitions is not surprising. Such a preference is strong when both \(j\) values involved in the transitions are large with respect to nuclear spin \(I\), while it becomes weak in the opposite case. A less clear minor feature lies in the fact that, for fixed values of \(j\), the preference is minimum when \(|\Delta j| = 3\).

REFERENCES

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SUPPORTING INFORMATION

Additional Supporting Information may be found in the online version of this article:

Data tables. Rates of spinless rotational transitions, rates for quasi-elastic purely hyperfine transitions and hyperfine propensities.

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