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HD molecules at high redshift: cosmic ray ionization rate in the diffuse interstellar medium

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ABSTRACT
We present a systematic study of deuterated molecular hydrogen (HD) at high redshift, detected in absorption in the spectra of quasars. We present four new identifications of HD lines associated with known H2-bearing Damped Lyman-α systems. In addition, we measure upper limits on the HD column density in 12 recently identified H2-bearing DLAs. We find that the new HD detections have similar N(HD)/N(H2) ratios as previously found, further strengthening a marked difference with measurements obtained from analysis of H2 and associated C I lines. We are able to constrain the cosmic ray ionization rate (CRIR, \( \chi \)) for the new HD detections and for eight known HD-bearing systems where priors on UV flux (\( \chi \)) and number densities (\( n \)) are available. We find significant dispersion in \( \chi \), from a few \( \times 10^{-18} \) s\(^{-1}\) to a few \( \times 10^{-15} \) s\(^{-1}\). We also find that \( \chi \) strongly correlates with \( n \) – showing almost quadratic dependence, slightly correlates with Z, and does not correlate with \( n \), which probably reflects a physical connection between cosmic rays and star-forming regions.

Key words: cosmic rays – ISM: molecules – galaxies: ISM – quasars: absorption lines.

1 INTRODUCTION
The formation and evolution of galaxies are intimately linked to their interstellar medium (ISM). Indeed, the ISM provides the fuel for star formation and in turn, the physical and chemical properties of the ISM are affected by stars (through UV radiation, cosmic rays, winds, enrichment by metals and dust, mechanical energy injection, etc.). The ISM presents several phases: the cold dense phases (cold neutral medium, CNM, itself including molecular phases) that may eventually collapse to form stars, the warmer, less dense phases (warm neutral and ionized medium, WNM and WIM, respectively), and the hot ionized medium (HIM) (Field, Goldsmith & Habing 1969; McKee & Ostriker 1977). These phases are well studied in the local Universe via analysis of emission over a large range of wavelengths in the electromagnetic spectrum from X-ray (Snowden et al. 1997) to radio (Heiles & Troland 2003), but their description through the Galaxy. This is likely due to differences in physical conditions and metallicity between the local and the high-redshift Universe via analysis of emission over a large range of wavelengths in the electromagnetic spectrum from X-ray (Snowden et al. 1997) to radio (Heiles & Troland 2003), but their description is still limited at high redshift, due to flux dimming at cosmological distances and significantly coarser spatial resolution available for emission-line studies of most tracers of the ISM.

This problem can be overcome by absorption-line spectroscopy. Both the WNM and CNM at high redshift are detectable in the spectra of background quasars and \( \gamma \)-ray burst (GRB) afterglows as Damped Lyman-α systems (DLAs) – absorption-line systems with the highest column densities of neutral hydrogen, \( \log N(H I) > 20.3 \)1) and a collection of associated metal lines (for a review, see Wolfe, Gawiser & Prochaska 2005). Most DLAs actually represent WNM (Srianand et al. 2005; Neeleman, Prochaska & Wolfe 2015), while CNM is much more rarely detected (in a few per cent of DLAs, see e.g. Balashev & Noterdaeme 2018).

One of the main tracers of CNM is molecular hydrogen (H2), the most abundant molecule in the Universe. Using UV absorption lines of H2 in the Lyman and Werner bands, one can probe diffuse and translucent molecular clouds along the line of sight (Ledoux, Petitjean & Srianand 2003; Noterdaeme et al. 2008, 2010; Balashev et al. 2017; Ranjan et al. 2018). If the H2 column density is large enough, the less abundant isotopologue, HD, can also be detected (Varshalovich et al. 2001). To date, HD lines have been detected only in 12 intervening systems among \( \sim 40 \) confirmed H2-bearing DLAs at high redshift (\( z > 0 \)) (Noterdaeme et al. 2008; Balashev, Ivanchik & Varshalovich 2010; Ivanchik et al. 2010; Noterdaeme et al. 2010; Tumlinson et al. 2010; Ivanchik et al. 2015; Klimenko et al. 2015; Klimenko et al. 2016; Balashev et al. 2017; Noterdaeme et al. 2017; Kosenko & Balashev 2018; Rawlins et al. 2018). This number remains limited since the detailed analysis of H2 and HD lines can be done only in high-resolution quasar spectra, which require observations with the largest optical telescopes. Also, as mentioned before, the incidence rate of the cold ISM in DLAs at high \( z \) is quite low. Hence, blind searches for HD/H2 are very inefficient (Jorgenson et al. 2014). Notwithstanding, in recent years several

1Here and in what follows, N is the column density in cm\(^{-2}\).

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efficient techniques were proposed to pre-select saturated H₂ lines in DLAs where HD is then easier to detect (Balashev et al. 2014; Ledoux et al. 2015; Noterdaeme et al. 2018). Some of the high-redshift N(H₂)/2N(H₂) measurements lie close to the primordial isotopic (D/H₂) ratio, triggering the discussion on whether the molecular isotopic ratio could serve as a proxy for D/H, in particular at high column densities where the cloud is thought to be fully molecularized (e.g. Ivanchik et al. 2010). However, models suggest that the HD/H₂ ratio varies significantly with depth into the clouds (Le Petit, Roueff & Le Bourlot 2002; Liszt 2015; Balashev & Kosenko 2020) since HD and H₂ have different main formation mechanisms: H₂ is forming mainly on the surface of dust grains, while HD is mostly formed via fast ion-molecular reactions. At the same time, destruction of both HD and H₂ mainly occurs via photodissociation by UV photons. This implies that the HD/H₂ ratio is sensitive to a combination of physical conditions, and that the HD/H₂ ratio can differ from the isotopic ratio even at high column densities in self-shielded regions (Balashev & Kosenko 2020). Moreover, under some conditions the D/HD transition may take place earlier than the H/H₂ transition (Balashev & Kosenko 2020), which leads to HD/2H₂ > D/H (Tumlinson et al. 2010; Noterdaeme et al. 2017), and therefore HD/H₂ may not be used as a lower limit for the isotopic ratio.

From the known HD-bearing systems, it was found that the relative HD/H₂ abundance tends to systemically be higher at high redshift than in the Galaxy (Snow et al. 2008; Balashev et al. 2010; Tumlinson et al. 2010; Ivanchik et al. 2015). This discrepancy cannot be solely explained by the progressive destruction of deuterium, since the astration of D through stellar evolution is expected to be small (Dvorkin et al. 2016). Therefore, the most probable explanation is to be sought in the differences in physical conditions between the ISM of the Galaxy and that of distant galaxies. Indeed, the models of ISM chemistry show that the HD/H₂ ratio is sensitive to the physical conditions in the ISM – UV flux, cosmic ray ionization rate (CRIR), metallicity, number density, and cloud depth (van Dishoeck & Black 1986; Le Petit et al. 2002; Čirković, Damjanov & Lalović 2006; Liszt 2015; Balashev & Kosenko 2020). Among these parameters, the CRIR seems to play a major role, being extremely important for the ISM chemistry. Indeed, cosmic rays are an important source of heating and one of the main ionizing sources and therefore drive almost all of the chemistry in the ISM. In the case of HD, cosmic rays promote the main channel of its formation as follows:

$$\text{H}^\text{CR} \rightarrow \text{H}^+ \rightarrow \text{D} \rightarrow \text{H}_2 \rightarrow \text{HD}$$  \hspace{1cm} (1)

Therefore, HD can, in principle, be used to constrain the CRIR (e.g. Black & Dalgarno 1973; Watson 1973; Hartquist, Doyle & Dalgarno 1978; Federman, Weber & Lambert 1996; Liszt 2015; Balashev & Kosenko 2020). Such an independent constraint would be extremely valuable, given the still loose constraints on CRIR in both the local Universe (see e.g. Hartquist et al. 1978; van Dishoeck & Black 1986; Federman et al. 1996; Indriolo et al. 2007; González-Alfonso et al. 2013, 2018; van der Tak et al. 2016; Neufeld & Wil进行全面的关键词搜索。例如，我们通过将HD作为CRIR的探针并考虑其在探测高红移DLAs中的稀有性，我们进行了一次系统性的搜索。首先，我们在历史上已经报道的DLA中进行搜索。然后，在高红移DLA上对CRIR进行了限制。最后，我们研究了在不同物理参数下的HD行为，以确保能够得到一致的结论。
### Table 1. H$_2$-bearing DLA systems searched for HD.

| Quasar      | $z_{em}$ | $z_{abs}$ | log $N$(HI) | [X/H]$^a$ | X   | log $N$(H$_2$) | References$^b$ |
|-------------|----------|-----------|-------------|----------|-----|----------------|----------------|
| J0136+0040  | 2.78     | 2.779     | 20.73 ± 0.01| −0.58 ± 0.03| S   | 18.6 ± 0.08 | 1               |
| J0858+1749  | 2.65     | 2.625     | 20.40 ± 0.01| −0.63 ± 0.02| S   | 19.7 ± 0.07 | 1               |
| J0906+0548  | 2.79     | 2.567     | 20.13 ± 0.01| −0.18 ± 0.05| S   | 18.88 ± 0.02 | 1               |
| J0910+0154  | 2.18     | 2.107     | 20.75 ± 0.04| 0.17 ± 0.07 | Zn  | 20.11 ± 0.06 | 2, 3            |
| J0946+1216  | 2.66     | 2.607     | 21.15 ± 0.02| −0.48 ± 0.01| S   | 19.97 ± 0.01 | 1               |
| J1143+1420  | 2.58     | 2.323     | 21.64 ± 0.06| −0.80 ± 0.06| Zn  | 18.3 ± 0.1   | 4               |
| J1146+0743  | 3.03     | 2.840     | 21.54 ± 0.01| −0.57 ± 0.02| Zn  | 18.82 ± 0.02 | 1               |
| J1236+0010  | 3.02     | 3.033     | 20.78 ± 0.01| −0.58 ± 0.04| S   | 19.76 ± 0.01 | 1               |
| J1215+0352  | 2.68     | 2.46      | 21.83 ± 0.01| −0.84 ± 0.23| Zn  | 21.31 ± 0.01 | 5               |
| J2232+1242  | 2.30     | 2.230     | 21.75 ± 0.03| −1.48 ± 0.05| Zn  | 18.56 ± 0.02 | 4               |
| J2347−0051  | 2.62     | 2.588     | 20.47 ± 0.01| −0.60 ± 0.06| S   | 19.44 ± 0.01 | 1               |

### Table 2. Known HD-bearing DLA systems.

| Quasar      | $z_{em}$ | $z_{abs}$ | log $N$(HI) | [X/H]$^a$ | X   | log $N$(H$_2$) | N(HD) | References$^b$ |
|-------------|----------|-----------|-------------|----------|-----|----------------|-------|----------------|
| J0000+0048  | 3.03     | 2.5255    | 20.8 ± 0.1  | 0.46 ± 0.45 | Zn  | 20.43 ± 0.02 | 16.64 ± 0.16 | 1               |
| B0120−28    | 0.434    | 0.18562   | 20.50 ± 0.10| −1.19 ± 0.15| S   | 20.00 ± 0.10 | 14.82 ± 0.15 | 2               |
| Q0528−2505  | 2.77     | 2.81112   | 21.35 ± 0.10| −0.68 ± 0.02| Zn  | 17.85 ± 0.02 | 13.33 ± 0.02 | 3, 4            |
| J0643−5041  | 3.09     | 2.658601  | 21.03 ± 0.08| −0.91 ± 0.09| Zn  | 18.54 ± 0.01 | 13.65 ± 0.07 | 5               |
| J0812+3208  | 2.7     | 2.624433  | 21.35 ± 0.10| −0.81 ± 0.10| Zn  | 19.93 ± 0.04 | 15.71 ± 0.07 | 6, 7            |
| J0843+0221  | 2.92     | 2.786     | 21.82 ± 0.11| −1.52 ± 0.08| Zn  | 21.21 ± 0.02 | 17.35 ± 0.15 | 8               |
| J1254+0815  | 2.57     | 2.3377    | 20.90 ± 0.08| −1.32 ± 0.12| S   | 19.57 ± 0.10 | 15.53 ± 0.17 | 9, 10           |
| J1237+0647  | 2.78     | 2.68959   | 20.00 ± 0.15| 0.34 ± 0.12 | Zn  | 19.20 ± 0.13 | 14.48 ± 0.05 | 11              |
| J1331+170   | 2.08     | 1.77670   | 21.18 ± 0.04| −1.22 ± 0.10| Zn  | 19.43 ± 0.10 | 14.83 ± 0.15 | 6, 12           |
| J1439+1117  | 2.58     | 2.41837   | 20.10 ± 0.10| 0.16 ± 0.11 | Zn  | 19.38 ± 0.10 | 14.87 ± 0.03 | 13, 14          |
| J2100−0641  | 3.14     | 3.09149   | 21.05 ± 0.15| −0.73 ± 0.15| Si  | 18.76 ± 0.04 | 13.83 ± 0.06 | 15, 16          |
| J2103−0050  | 2.261    | 2.0593    | 19.18 ± 0.15| −0.19 ± 0.10| S   | 17.94 ± 0.01 | 13.87 ± 0.06 | 17              |
| J2340−0053  | 2.083    | 2.05      | 20.35 ± 0.05| −0.52 ± 0.06| S   | 18.6 ± 0.02 | 14.11 ± 0.06 | 6               |

Notes:

$^a$ Metallicity with respect to solar (Asplund et al. 2009): $[\text{X/H}] = \log \left( \frac{N(\text{X})}{N(\text{H})} \right)$.  
$^b$ References: (1) Noterdaeme et al. (2017), (2) Oliveira et al. (2014), (3) Klimenko et al. (2015), (4) Balashev et al. (2020), (5) Albornoz Vázquez et al. (2014), (6) Balashev et al. (2010), (7) Jorgenson et al. (2009), (8) Balashev et al. (2017), (9) Ivan'chik et al. (2010), (10) Balashev et al. (2011), (11) Noterdaeme et al. (2010), (12) Carswell et al. (2011), (13) Srianand et al. (2008), (14) Noterdaeme et al. (2008), (15) Ivan'chik et al. (2015), (16) Jorgenson et al. (2010), (17) Klimenko et al. (2016), (18) Rawlins et al. (2018).

$^c$ This work.
3 ANALYSIS

We analysed the absorption lines using multicomponent Voigt profile fitting. The unabsorbed continuum was typically constructed by eye using spline interpolation constrained by the regions free from any evident absorption lines (see e.g. Balashev et al. 2019). The lines were fitted simultaneously and the spectral pixels that were used to constrain the model were selected by eye to avoid blends (mainly with Ly α forest lines). The best value and interval estimates on the fitting parameters (Doppler parameter, b, column density, \(N\) and redshift, \(z\)) were obtained with a Bayesian approach, using standard \(\chi^2\) likelihood to compare the data and the model. To sample the posterior distribution function of the parameters we used Monte Carlo Markov Chain (MCMC) (see e.g. Balashev et al. 2017) with affine-invariant sampling (Goodman & Weare 2010). By default the priors on most parameters were assumed to be flat (for \(b\), \(N\) and \(z\)). However, for most X-shooter spectra, the resolution is not high enough to accurately resolve the velocity structure and some HD lines can be in the saturated regime. In these cases, we found that the column densities and Doppler parameters can be highly degenerated, resulting in uncertain constraints. Therefore, we used priors on the number of components, their redshifts and Doppler parameters from the analysis of \(H_2\) or C I absorption lines (see e.g. Balashev et al. 2019). This is adequate, since \(H_2\) is usually constrained by a large number of lines (\(\sim 50-100\)) and C I is fitted in the region out of Ly α forest. We used mostly components where the column density of \(H_2\) exceeds \(\log N(H_2) \gtrsim 18\), since for the lower \(H_2\) columns the expected HD column densities will be much lower than what the data can constrain, i.e. even upper limits will be uninformative.

Moreover, we found that in the X-Shooter spectra, the continuum placement for some HD lines is non-trivial. We estimated the resulting uncertainty independently using the following procedure. We performed a large number (\(\sim 500\)) of realizations, where we randomly shifted the continuum level for each line. The values of the shifts were drawn from a normal distribution with dispersion corresponding to the mean uncertainty of spectral pixels at the positions of the absorption lines. For each realization, we also randomly drew an HD Doppler parameter using the constraints obtained from \(H_2\). The redshift uncertainty from \(H_2\) (or C I) in the most cases is quite low and has only marginal effect on the results. We then fitted each realization \(i\) with fixed \(b\) and \(z\) and obtained the best-fitting column density \(N(\text{HD})\). We obtained the final HD column density measurement from the distribution of \(N(\text{HD})\). We found that the uncertainties on HD column densities increase in the most cases by a factor of \(\sim 2\) compared to the MCMC fit with fixed continuum, meaning that the continuum placement uncertainty contributes significantly to the total \(N(\text{HD})\) uncertainty budget at the medium resolution.

We summarize the results of fitting HD lines in Table 3 and provide specific comments on each system as follows:

3.1 VLT/X-shooter data

3.1.1 J0136+0440

We only tentatively detected HD absorption lines at the expected position based on the redshift of the main \(H_2\) component (\(z = 2.779430\)) with the column density \(\log N(H_2) = 18.64^{+0.08}_{-0.09}\) and Doppler parameter \(b = 7.7^{+2.4}_{-1.9}\) km s\(^{-1}\). Therefore, fixing \(z\) and using prior on Doppler parameter from \(H_2\) analysis, we placed only an upper limit to the HD column density in this component, \(\log N(\text{HD}) < 14.5\). The fits to the unblended HD absorption lines are shown in Fig. A1. Here in the following figures we show only those HD absorption lines that are not totally blended with other absorption lines (from Ly α forest and/or \(H_2\) and metal lines from corresponding DLA).

3.1.2 J0858+1749

We detected HD absorption lines at the position of \(H_2\) component (\(z = 2.62524\)) that has log \(N(H_2) = 19.72^{+0.01}_{-0.02}\) and \(b = 7.9^{+0.4}_{-0.3}\) km s\(^{-1}\). To fit HD lines we fixed \(z\) and used \(b\) as a prior from \(H_2\) analysis. Using the HD L8-0R(0) line and red wings of HD L4-0R(0), HD L7-0R(0), HD L11-0R(0), and HD L12-0R(0) absorption lines (see Fig. A2), we constrained \(\log N(\text{HD}) = 14.87^{+0.06}_{-0.09}\).

3.1.3 J0906+0548

We only tentatively detected HD absorption lines at the position of the main \(H_2\) component (\(z = 2.56918\)) that has \(\log N(H_2) = 18.87^{+0.02}_{-0.01}\) and \(b = 6.3^{+0.1}_{-0.1}\) km s\(^{-1}\). Although we did find HD lines at the expected positions, all of them are partially or fully blended with other absorption lines (see Fig. A3). Therefore, using \(z\) and prior on \(b\) obtained from \(H_2\) analysis, we were only able to place an upper limit to the HD column density in this component to be \(\log N(\text{HD}) < 14.7\).

3.1.4 J0917+0154

This system was selected by Ledoux et al. (2015) in their search for the cold gas at high redshift through C I lines. The detection and analysis of \(H_2\) was presented by Noterdaeme et al. (2018) (they reported the total column density \(N(H_2) = 20.11^{+0.06}_{-0.06}\) and the metal lines were studied by Zou et al. (2018). Unfortunately, due to low resolution and relatively high velocity extent of \(H_2\) lines, almost all HD lines are blended, including usually available L3-0R0, L4-0R0, and W0-0R0 lines. The only not blended line L0-0R0 has a very low oscillator strength and therefore we were able to put only very conservative upper limit on the HD column density using priors on the redshifts and Doppler parameters for three components fit obtained from the refitting jointly C I and \(H_2\) absorption lines. The fit to C I and HD lines is shown in Fig. A4 and H2 lines profiles are presented in Fig. A18. The detailed fit result is given in Table A1.

3.1.5 J0946+1216

The detection of HD at the position of the main \(H_2\) component (\(z = 2.60642\), log \(N(H_2) = 19.96^{+0.01}_{-0.02}\), \(b = 9.8^{+0.8}_{-0.8}\) km s\(^{-1}\)) for this system is also tentative. Unfortunately, the spectrum is very noisy and significantly contaminated by highly saturated \(H_2\) lines and intervening Ly α forest absorption. Therefore, we fixed \(z\) and used Doppler parameter from \(H_2\) analysis as a prior. Hence, we were only able to obtain relatively loose constraint on the HD column density in this component to be \(\log N(\text{HD}) < 15.2\), see Fig. A5.

3.1.6 J1143+1420

This extremely saturated DLA at \(z = 2.3228054\) was previously analysed by Ranjan et al. (2020) and \(H_2\) column density was found to be \(\log N(H_2) = 18.3^{+0.1}_{-0.1}\). We looked for HD lines associated with \(H_2\), and we were able to place an upper limit on HD column density.
We used fixed $z$ and prior on Doppler parameter from $H_2$ analysis, and got $N(\text{HD}) < 15$. The fit to HD lines is shown in Fig. A6.

### 3.1.7 J1146+0743

We do not detect HD absorption lines at the position of both $H_2$ components ($z = 2.84163$ and $2.83946$ with $N(H_2) = 18.76 \pm 0.1$ and $17.94^{+0.11}_{-0.13}$, respectively). Therefore, we constrained log$N(\text{HD}) < 14.4$ and log$N(\text{HD}) < 14.5$ for the red and blue components, respectively, using a combination of $\text{HD} L3-0R(0),\text{HD} L8-0R(0),\text{HD} W0-0R(0),\text{HD} W1-0R(0),\text{HD} L11-0R(0)$, and $\text{HD} L12-0R(0)$ lines and priors on $b$ and fixed $z$ from $H_2$ analysis. The spectrum at the expected positions of HD absorption lines is shown in Fig. A7.

### 3.1.8 J1236+0010

We do not detect HD absorption lines at the position of $H_2$ component of DLA ($z = 3.03292$, log$N(H_2) = 19.76 \pm 0.01$, $b = \ldots$)
$2.06678$. Knowing that C I is an excellent tracer of H$_2$ in ISM (Noterdaeme et al. 2018), we searched for H$_2$ and HD molecules in this system as well. We used the Keck/HIRES spectrum whose reduction is detailed in Balashev et al. (2010). We detected HD$_2$ absorption lines from $J < 4$ rotational levels, which we fitted using a one-component model, with tied redshifts and Doppler parameter rotational levels for all levels. Indeed, H$_2$ lines are located at the blue end of the spectrum, covering only one-two unblended H$_2$ lines from each rotational level. The fit results are given in Table A2 and line profiles are shown in Fig. A19. Using relative population of $J = 1$ and $J = 0$ levels, we found the excitation temperature to be $T_{\text{eff}} = 67^{+4}_{-3}$ K.

Unfortunately, only two HD lines (L0-0R0 and L1-0R0) were covered in this spectra and only the weakest HD L0-0R0 line from this system was unblended (see Fig. A19). Thus, we estimated only an upper limit to the HD column density, fixing the redshift and Doppler parameter from H$_2$, and obtained log $N$(HD) < 14.4.

### 3.2.3 J0816+1446

The multicomponent H$_2$-bearing DLA system towards J0816+1446 was identified by Guimarães et al. (2012). This system has quite large redshift and hence is significantly blended with the Ly $\alpha$ forest lines. Guimarães et al. (2012) reported H$_2$ in two components, with one at $z = 3.28742$ indicates a significantly high H$_2$ column density, log $N$(H$_2$) = 18.66 $\pm$ 0.27 to be searched for HD. We refit H$_2$ absorption lines at $z = 3.28742$ with three subcomponents, since it provides a better fit, and measured the total log $N$(H$_2$) = 18.51 $\pm$ 0.04 in agreement with Guimarães et al. (2012). Unfortunately, all HD lines are blended and therefore using fixed $z$ and Doppler parameter from H$_2$ analysis we were able to obtain an upper limits on the HD column densities log $N$(HD) $\lesssim 15$ from the L4-0R(0) line (fit results are presented in Table A3 and Fig. A13).

### 3.2.4 J1311+2225

This multicomponent H$_2$-bearing DLA system was selected through C I by Ledoux et al. (2015). Noterdaeme et al. (2018) reported log $N$(H$_2$) = 19.69 $\pm$ 0.01 in this system, using a single-component model, but they noted that four components for H$_2$ lines can be distinguished. We refit H$_2$ and C I lines in this system using four-component model. First, we fit C I absorption lines from three fine-structure levels, where we tied Doppler parameters for each component. Then we performed a four-component fit to the H$_2$ lines, where the selection of initial guess of components was based on C I result. For H$_2$, we tied Doppler parameters only between $J = 0$ and $J = 1$ levels, while Doppler parameters for other rotational levels were allowed to vary independently. However, since the components are significantly blended among themselves and the data are quite noisy, we added two penalty functions to the likelihood. The first one is set to artificially suppress situations where the Doppler parameter of the some J level would be lower than that of the $J-1$ level. This is well motivated physically and observationally, since the increase of the Doppler parameters for the higher H$_2$ rotational levels has been established in many H$_2$ absorption systems (see e.g. Lacour et al. 2005; Noterdaeme et al. 2007; Balashev, Varshalovich & Ivanchik 2009). The other penalty is to keep a reasonable excitation diagram of H$_2$: we penalized models with $T_{\text{eff}} < J - 1 > T_{\text{eff}, J+1}$. This is also

\[T_{\text{eff}, J+1} \text{ is the excitation temperature between } J \text{ and } J+1 \text{ levels} \]
reasonably motivated by both observations and modelling (see e.g. Klimenko & Balashev 2020). Therefore, we get total H$_2$ column density to be N(H$_2$) = 19.59 ± 0.01, which is a bit lower than the value 19.69 ± 0.01 reported previously (Noterdaeme et al. 2018). The fitting results are shown in Table A4 and C I and H$_2$ profiles in Figs A20, A21, A22, A23, and A24.

We also estimated metallicity in this system. Unfortunately, very few metal lines, which are usually used to obtain metallicity, were covered in this spectrum, and almost all covered lines are blended. Therefore, to obtain metallicity we used Zn II 2062 line. We fitted this line, assuming four components in the positions of C I components, and obtained Zn II total column density to be 12.84$^{+0.09}_{-0.11}$, therefore the metallicity is $-0.3^4_{-0.13}$ relative to solar. The fit to Zn II absorption line is shown in Fig. A22.

We again used a four-component model to analyse HD, associated with C I components. We found that component 3 for HD is shifted in comparison with C I lines. However, component 3 in C I has quite large Doppler parameter, which indicates that there is velocity structure within this component, that meanwhile we cannot resolve due to low quality of the spectrum and mutual blending from other components. So for HD we did not use the H$_2$ and C I priors on redshifts (except weak component 1, where only upper limit on HD column density could be placed) and Doppler parameters. After the MCMC procedure we found HD to be detected in components 2, 3, and 4, and the redshifts of the components are well agree within uncertainties (see Table A4). Component 1 is too weak, so we could only place an upper limit on N(HD) there. We also tentatively detected absorption lines from HD J = 1 in this system. Using the joint fit HD J = 1 lines with Doppler parameters tied to HD J = 0 we placed upper limits on HD J = 1 column density in three components (we did not fit HD J = 1 lines in the bluest component, since HD column density is too low). Using N(HD J = 1)/N(HD J = 0) ratio we were able to make additional constraints on number densities in the medium (see e.g. Balashev et al. 2010; Liszt 2015) to be n $\lesssim$ 3.3 cm$^{-3}$, n $\lesssim$ 3.2 cm$^{-3}$, and n $\lesssim$ 3.7 cm$^{-3}$ in the components 2, 3, and 4, respectively. The fit to the HD lines is shown in Figs A14 and A15 and HD column densities are reported in Table A4.

3.2.5 J 2140−0321

H$_2$ absorption lines were previously found and analysed by Noterdaeme et al. (2015) and Ranjan et al. (2020) at z = 2.339 and H$_2$ column density was found to be quite large log N(H$_2$) = 20.13. To fit HD absorption lines we used together the spectra, obtained by X-shooter and UVES. However, since the UVES spectrum is very noisy, and X-shooter is low-resolution hence it is not appropriate for HD analysis. Therefore, we were able only to place upper limit on HD column density to be log N(HD) $< 14.6$ using the priors on the Doppler parameters and the redshifts obtained from H$_2$ analysis (Noterdaeme et al. 2015), see Fig. A16.

3.2.6 J 2340−0053

C I and H$_2$ absorption lines in the DLA at z $\approx$ 2.055 towards J 2340−0053 were first reported by Jorgenson et al. (2010). These authors found C I in nine components, while they fitted H$_2$ using a six components model. This spectrum was recently reanalysed by Rawlins et al. (2018) with seven components for both C I and H$_2$ and found their redshifts to be consistent with each other. HD absorption lines, associated with H$_2$, were later independently detected by Kosenko & Balashev (2018) and Rawlins et al. (2018). In this paper, we present a detailed reanalysis of HD, H$_2$, and C I absorption lines.

Using the reduced 1D-spectrum of J 2340−0053 from the KO-DIAQ data base (O’Meara et al. 2017), we refitted C I, H$_2$, and HD absorption lines with seven component model using the same methodology as in the previous section for J 1311+2225. We fit C I lines first, taking into account the partial coverage of the background emission-line region by C I line at z $\approx$ 1560 Å reported by Bergeron & Boissé (2017). We fit the covering factors as an independent parameter following the methodology from Balashev et al. (2011). We found that a fit with three independent covering factors for each of the three main components provides a better fit, than using a single covering factor for all components. We then used the C I fit as first guess to the redshifts of the H$_2$ lines. Unfortunately, three central components are significantly blended with each other in almost all H$_2$ absorption lines from J = 0, 1, 2, and 3 rotational levels. Therefore, we used the redshifts determined during C I fit as priors, and as for J 1311+2225, we used penalty functions during H$_2$ analysis to reproduce physically reasonable constraints. We obtained the total H$_2$ column density to be log N(H$_2$) = 18.57 ± 0.02, which is higher than reported by Rawlins et al. (2018), log N(H$_2$) = 17.99 ± 0.05. The difference is partly due to the fact that Rawlins et al. (2018) tied all H$_2$ Doppler parameters for J > 0 to H$_2$ J = 0, while we tied only H$_2$ J = 1 and allowed increasing b-values for other levels. The fitting results are shown in Table A5 and C I and H$_2$ profiles in Figs A26, A27, A28, and A29.

We fit HD absorption lines at the positions of these components using the priors on the Doppler parameters from the fit of J = 0 and J = 1 rotational levels of H$_2$. However, the exact b-values affect little the results since the HD absorption lines are optically thin. The obtained total HD column density is log N(HD) = 14.11 ± 0.06, which is a bit lower than found by Rawlins et al. (2018) log N(HD) = 14.28 ± 0.08. The fit to the HD lines is reported in Table A5 and shown in Fig. A17.

4 RESULTS

We summarize our new measurements of the HD (and H$_2$) column densities and the relative abundances of N(HD)/2N(H$_2$) in Table 3. In total, we report four new detections of HD molecules in high-redshift DLAs (sometimes in several components) and place upper limits for other 12.

Fig. 1 compares the HD and H$_2$ column densities in the Galaxy (Snow et al. 2008) and at high redshift (new measurements and values from Table 2). We also compare the data to the primordial D/H isotopic ratio derived from the updated big bang Nucleosynthesis (BBN) calculations (Pitrou et al. 2018) and $\Omega_b h^2$ from Planck Collaboration VI (2020). One can see that the molecular ratios are well below the primordial isotopic ratio in the Galaxy, while distant measurements do not show such a tendency and instead indicate a systematically higher HD/H$_2$ relative abundance than locally at specific log N(H$_2$), and closer to the BBN value.

Several processes can in principle affect the HD/H$_2$ relative abundance, such as fractionation and astration of deuterium. The chemical fractionation of D (the process through which D efficiently replaces hydrogen in complex molecules, such as D$_2$, HDO, D$_2$O, NH$_2$D, NH$_2$D$^+$, ND$_3$, D$_2$O$^+$, etc.) should play a minor role, since complex molecules mainly reside in the cold dense medium with n$_D$ $\gtrsim$ 10$^3$ cm$^{-3}$ and T $\lesssim$ 25 K (see e.g. Kim et al. 2020, and references therein), which is not the case here as we probe more diffuse clouds. Measurements at high z tend to probe lower metallicities than locally and hence represent gas that has been...
Figure 1. Relative abundance of HD and H$_2$ molecules. The green circles, red squares, and yellow triangles correspond to known HD-bearing systems at high redshift (for references, see text), new HD detections at high redshift and upper limits on HD column densities (filled and empty squares; this work), and measurements in the Galaxy (Snow et al. 2008), respectively. The solid blue line shows the D/H isotopic ratio estimated using standard BBN calculations and $\Omega_b h^2$ from the Planck Collaboration VI (2020).

least processed in stars. Such gas should therefore be less affected by astration of deuterium than measurements in the Galaxy (i.e. $\sim$10 Gyr later). However, Dvorkin et al. (2016) showed that D/H is never reduced to less than one-third of its primordial ratio, i.e. astration cannot explain the observed discrepancy. On the other hand, the low metallicity affects the HD abundance from the chemical pathway (Liszt 2015; Balashev & Kosenko 2020). Indeed, the lower the metallicity, the lower the abundances of dust and carbon (and therefore the amount of electrons released by C; in our model we assume that the rate of grain-assisted recombination and the carbon abundance are scaled linearly with metallicity). This results in a drop of H$^+$ radiative (as the electron fraction decreases) and grain-assisted recombination rates at low metallicity. Hence, the fractions of ionized hydrogen and deuterium are high at low metallicity, owing to D$^+$ being mainly produced by charge exchange with H$^+$. This then results in an increased HD formation rate through the reaction:

\[ \text{H}_2 + \text{D}^+ \rightarrow \text{HD} + \text{H}^+ \quad (2) \]

The enhanced HD formation rate consequently increases the HD abundance relative to H$_2$. Interestingly, in certain physical conditions, this may lead to a D/HD transition occurring earlier (lower penetration depth) in ISM clouds than the H/H$_2$ transition (Balashev & Kosenko 2020). The evident observational consequence of this is that $N(\text{HD})/2N(\text{H}_2) > \text{D/H}$, while the opposite case was generally obtained (e.g. Le Petit et al. 2002), since naively HD is always significantly less self-shielded in the medium than H$_2$. In conclusion, the typically lower metallicities at high z can in principle explain the systematic difference in relative HD/H$_2$ abundance between high-z and Milky-Way measurements (see also Liszt 2015).

5 PHYSICAL CONDITIONS

The relative HD/H$_2$ abundance depends not only on the metallicity, but also on the physical conditions in the medium – number density, UV flux, and CRIR (Le Petit et al. 2002; Cirković et al. 2006; Liszt 2015). To describe this dependence, we used recently published simple semi-analytic description of the dependence of the HD/H$_2$ ratio on these parameters (Balashev & Kosenko 2020). This method includes solving the HD balance equation between formation and destruction processes in a plane-parallel, steady-state cloud and permits the determination of how $N(\text{HD})$ – as a function of $N(\text{H}_2)$ – depends on the physical properties in the cloud, namely CRIR per hydrogen atom (CRIR, $\zeta$), UV field intensity (relative to Draine field, Draine 1978, $\chi$), number density ($n = n_{\text{HI}} + 2n_{\text{H}_2}$), and metallicity (Z). We assumed that the D/H isotopic ratio is $2.5 \times 10^{-5}$ for all systems, i.e. we neglected a possible astration of D, which is typically much smaller (Dvorkin et al. 2016) than the uncertainties of this method (see below).

To constrain the distributions of the physical parameters from the measured $N(\text{HD})$ and $N(\text{H}_2)$, we followed a Bayesian approach using affine-invariant Markov Chain Monte Carlo (MCMC) sampling (Foreman-Mackey et al. 2013). Because we do not have access to the total hydrogen ($N(\text{H}_1) + N(\text{H}_2)$) in individual HD-bearing components, the metallicity for each cloud was set to the overall metallicity in the corresponding DLA, as provided in Tables 1 and 2.\footnote{For simplicity, in our formalism, the carbon abundance (which is important for the ionization fraction), the H$_2$ and HD formation rates on dust grains

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Figure 2. Posterior probability functions for CRIR ($\zeta$), UV field intensity ($\chi$), and number density ($n$) obtained from HD/H$_2$ fitting for the system at $z = 2.625241$ towards J0858$+$1749. The diagonal panels show 1D marginalized posterior functions, non-diagonal show 2D posterior functions, where the dark- and light-blue regions correspond to 1$\sigma$ and 2$\sigma$ confidence levels, respectively.

6 DISCUSSION

We find the CRIRs to vary significantly from $\zeta \sim 10^{-18}$ to $10^{-15}$ s$^{-1}$, possibly reflecting a wide range of environments being probed by our sample. Indeed, DLA systems are selected owing to their absorption cross-section and likely probe the overall galaxy population, with a high fraction of low-mass galaxies at high redshift (e.g. Cen 2012), in which the star-formation and CRIRs are expected to vary significantly. Even though the HD/H$_2$ absorption systems in our sample do not necessarily probe the immediate environments of star formation, as we will show below, the measured high CRIR values correlate with the relatively high UV fluxes that reach up to 10 times the Draine field.

We find that the range of CRIR estimates are in line with other recent measurements both at high redshift (Muller et al. 2016; Shaw et al. 2016; Indriolo et al. 2018) and in nearby galaxies (van der Tak et al. 2016; González-Alfonso et al. 2013, 2018), which also show quite large dispersion. This dispersion can be partly due to the use of various methods, or connected to a real physical dispersion of the CRIR. Indeed, the measurements in the Galaxy (for a review see Padovani et al. 2020, and references therein) and in the lensed system at $z \sim 0.89$ towards PKS 1830$-$211 (Muller et al. 2016) show that this parameter can vary significantly between different sightlines even inside a given galaxy, mostly depending on the proximity to the CR accelerator. Le Petit et al. (2016) also present evidence of CRIR enhancement in the centre of the Galaxy relative to the disc. Finally, we think that the comparison of previous data with our measurement is likely not straightforward since different methods have been used, which probe various environments. Indeed, the aforementioned and most recent constraints on CRIR in local and high-$z$ galaxies have been based on oxygen-bearing species (OH$^+$ and H$_2$O$^+$). Since these have been analysed in quite luminous starburst galaxies with roughly solar metallicity and high star formation rates, they may sample rather high CRIR values compared to the overall galaxy population.

Fig. 3 shows and compares our measurements with literature ones in the [$\zeta$, log N(H$_2$)] plane. An attenuation of the CRIR with increasing column density is theoretically expected (Padovani et al. 2009). However, we do not see strong evidence for a correlation between $\zeta$ and N(H$_2$) in our sample, probably because of the large dispersion (unweighted Pearson test gives correlation coefficient $r = -0.49$, with p-value 0.08). In addition, we probe mostly diffuse clouds with low cloud depths (except Q0843$+0221$ which will be discussed later), which may be insufficient to attenuate the cosmic ray flux. Additionally, the observed clouds should have quite large (N(H$_2$) $\gtrsim 10^{20}$ cm$^{-2}$) column densities of associated H$_2$, which is hard to constrain observationally, but which is also able to attenuate the CR flux, and therefore may provide an additional uncertainty in our calculations.

Previous measurements at high redshift and in the Galaxy show that in the case of a denser medium (e.g. dense cores, blue triangles, and the H$^+$ recombination on grains are assumed to scale linearly with the metallicity.
attenuated in the cloud at column densities $N$ throughout the cloud. However, it is expected that CRIR can be changed by available observations. The magnetic-field configuration in DLAs, which is not well probed by issues in the measurements themselves. However, the posterior distributions for individual systems (e.g. Fig 2) indicate that the measurements of $\zeta$ may correlate more strongly with $n$ than with $\chi$ (if any). We also note that the HD/H$_2$ ratio depends on the number density and metallicity (with similar sensitivity on variation of $n$ as for $\chi$, and even higher sensitivity for metallicity (see Balashev & Kosenko 2020); therefore, it is not evident why we should see a strong correlation between $\zeta$ and $\chi$, and a lack of correlation between $\zeta$ and $n$. This motivates us to assume that the $\zeta-\chi$ correlation has a real physical origin.

Indeed, we expect a common star-formation origin between cosmic rays and UV radiation. Furthermore, one can see that the slope of the log $\zeta$–log $\chi$ correlation is close to 2, i.e. CRIR increases quadratically with the strength of the UV field. This also may have reasonable explanation, since the low-energy cosmic rays ($\lesssim$100 MeV, which mostly determine the ionization rate) may have complex propagation behaviour, related to the diffusion in the ISM magnetic fields. In addition, taking into account the energy losses (see loss function for cosmic rays by Padovani et al. 2018), this may result in a local enhancement of the $\zeta/\chi$ ratio near the production sites and hence superlinear dependence of $\zeta$ on $\chi$, since UV photons escape much more easily from the star-forming regions.

Assuming that $\zeta \propto \chi^2$ dependence is real, we plot the $\sqrt{\zeta}/\chi$ as a function of log $N$(H$_2$) in Fig. 5. One can see that indeed for the main bulk of the systems with log $N$(H$_2$) in the range 18–20 the dispersion is significantly reduced and only within 1 dex, in comparison with the 3-dex dispersion of $\zeta$ in Fig. 3 (at the same time, we have not found a significant correlation of $\sqrt{\zeta}/\chi$ with neither $Z$ nor $n$). As already discussed, the single outlier value of $\sqrt{\zeta}/\chi$ with the highest log $N$(H$_2$) (corresponding to J0843+0221) can be related to cosmic

Figure 3. Estimated CRIR as a function of H$_2$ column density. Here, red squares are values obtained in this work, smaller squares are values obtained for other galaxies (blue – Muller et al. 2016; violet – Shaw et al. 2016; pink – Indriolo et al. 2018), and triangles are values measured in the Galaxy (yellow – Indriolo et al. 2007; light green are protostellar envelopes (for references, see table 6 from Padovani, Galli & Glassgold 2009); blue – Caselli et al. 1998; cyan – Shaw et al. 2008; brown – Mare & Bergin 2007; violet – Indriolo & McCall 2012; dark green – Indriolo et al. 2015).

Indeed, in our formalism, we assume CRIR, $\zeta$, to be constant throughout the cloud. However, it is expected that CRIR can be attenuated in the cloud at column densities $N$(H) $\gtrsim$ 10$^{20}$ cm$^{-2}$ (see e.g. Silsbee & Ivlev 2019). Therefore, if the CRIR is attenuated inside the cloud, then the derived value of $\zeta$ is lower than the incident value. That means that in principle, to draw accurate physical conclusions, cosmic ray propagation effects at high column densities should be taken into account properly. This would also require knowledge of the magnetic-field configuration in DLAs, which is not well probed by available observations.

Since the relative HD/H$_2$ abundance depends in opposite ways on $\zeta$ and $\chi$ (i.e. $N$(HD)/$N$(H$_2$) increases when $\zeta$ increases but also when $\chi$ decreases), the $\zeta-\chi$ correlation could be artificially introduced by issues in the measurements themselves. However, the posterior...
Figure 4. Estimated CRIR, $\zeta$, as a function of UV field strength (left-hand panel), number density (middle panel), and metallicity (right-hand panel), using the HD/H$_2$ relative abundance measured in DLAs at high redshift. The points are colour-coded by metallicity (left-hand and middle panels) and UV field strength (right-hand panel), with colour bars provided within each panel.

Figure 5. $\sqrt{\zeta/\chi}$ as a function of H$_2$ column density. The points are colour-coded by metallicity using the colour bar shown at the bottom.

ray propagation effects at high column densities, or its exceptionally low metallicity. In turn, at the lower column-density end, absorption systems probe very diffuse gas, where the application of the H/H$_2$ and D/HD transition model that we used should be taken with caution. The most important issue, in our opinion, is that the HD/H$_2$ ratio primarily depends on the hydrogen ionization fraction of the medium, while the CRIR was derived assuming that this ionization state of the diffuse ISM is mainly determined by CRIR and recombination on dust grains (Balashev & Kosenko 2020). In the case of a very diffuse medium, one can expect that the hydrogen ionization fraction is higher due to mixing with ionized and/or WNMs, the latter being mostly atomic and hence having higher ionization fractions. All this can effectively mimic the increase of the $\sqrt{\zeta/\chi}$ ratio at lower log $N$(H$_2$) values, which one can notice in Fig. 5. Additionally, the recombination rate coefficient can have a non-linear dependence on the metallicity (in our model, we assume it is linear), since it strongly depends on the properties of the dust and we have no strict constraints on it as a function of the metallicity. Indeed, a possible correlation of $\zeta$ with $Z$ (see right-hand panel of Fig. 4; excluding J0843+0221 as a possible outlier, we obtain a correlation coefficient of 0.67 with $p$-value 0.012) can be caused by this non-linear behaviour. Therefore, we caution to use a simple homogeneous model to estimate CRIR at low H$_2$ column densities. Otherwise, one can propose a physical explanation of the correlation of $\zeta$ with $Z$, as higher metallicity systems probe more massive galaxies (from well-known mass–metallicity relations, see e.g. Sanders et al. 2015), where star formation on average is expected to be higher than in low-mass galaxies and the cosmic ray flux (and therefore CRIR) is expected to be enhanced.

7 CONCLUSIONS

We have presented new measurements of HD molecules in high-$z$ absorption systems found in quasar spectra. We looked for HD in all known strong H$_2$-bearing systems and we detected HD molecules in four DLAs and placed upper limits on the HD column density for other twelve DLAs. So with this study, we significantly increased the sample of HD-bearing DLAs. We find that HD/H$_2$ relative abundances show large dispersion around the D/H isotopic ratio. In turn, observed HD/H$_2$ ratios can be used to estimate the gas physical conditions, in particular the CRIR (Balashev & Kosenko 2020). We find that the CRIR varies from a few $10^{-18}$ to a few $10^{-15}$ s$^{-1}$ in our sample of high-redshift absorbers, which likely reflects the wide range of environments and physical conditions probed by DLAs. These ranges and dispersion are also in line with previous measurements using various methods in the Galaxy as well as other galaxies.

We find that the CRIR is highly correlated with the UV field intensity in our sample, while it does not correlate with number density and only slightly with metallicity. These correlations suggest a physical connection between the sources of cosmic rays and those of UV radiation. Moreover, we find a quadratic dependence of $\zeta$ on $\chi$ in our sample, which is probably due to transport effects of low-energy cosmic rays. We caution however that these correlations may be artificial because of the dependence of $N$(HD)/$N$(H$_2$) on combination of $\zeta$, $\chi$, and $Z$.

Additionally, most of the methods currently used to determine CRIR involve a detailed chemical modelling of the regions related to the diffuse and translucent ISM. Being the dominant species in this region that determine the chemical network results, H$_2$ can be subject to strong systematics concerning the time-dependent chemistry, since the formation time-scale of H$_2$ can be relatively high in comparison with cloud lifetimes and steady-state models may not be appropriate (e.g. Balashev et al. 2010).
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DATA AVAILABILITY

The results of this paper are based on open data retrieved from the ESO and KECK telescope archives. These data can be shared on reasonable requests to the authors.

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SUPPORTING INFORMATION

Supplementary data are available at *MNRAS* online.

**Appendix A.** Profile fitting results.

**Appendix B.** 1d and 2d Posteriors on physical conditions.

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