INTRODUCTION

The formation of compact dusty clouds in diffuse interstellar medium (ISM) has been recently proposed and studied by Tsytovich et al. (2014). In the present paper, we examine the effect of the clouds on the rate of $H \rightarrow H_2$ transition in the ISM. We discuss the mechanisms leading to the formation of the clouds and the creation of gaseous clumps around them, analyze the main processes determining the efficiency of the recombination of atomic hydrogen on dust grains, and estimate the expected enhancement of the global $H_2$ formation due to the presence of the clouds. In conclusion, we argue that the compact dusty clouds provide optimum conditions for the atomic recombination in diffuse ISM, and point out some astrophysical implications of the resulting $H_2$ formation enhancement.

Subject headings: ISM: dust – plasmas – astrochemistry

1. INTRODUCTION

The formation of molecules at the surface of dust grains is a ubiquitous process occurring in various regions of interstellar medium (ISM). A prominent example is the formation of molecular hydrogen in diffuse ISM, a process of fundamental importance in astrophysics. Rate and timescales of $H_2$ formation in the diffuse ISM is a matter of debate. While classical estimates of the formation timescale made by Hollenbach et al. (1971) give a value of $\sim 3 \times 10^7$ yr (for the gas density of $\sim 10^2$ cm$^{-3}$), there is a strong evidence that this process should occur significantly faster (e.g., Hartmann et al. 2001; Mac Low & Klessen 2004; Liszt 2007; Mac Low et al. 2017).

The estimates for the reaction rate are based on a "natural" (and usually implicit) assumption that the ISM is homogeneous at (very) small spatial scales. However, there is a growing observational evidence that the gas density in diffuse ISM can vary substantially at scales below $\sim 1$ AU (Heiles 1997; Crawford 2002; Hartquist 2003; Heiles 2007; Stanimirović et al. 2010; Dutta et al. 2014). The importance of dense clumps in diffuse ISM was also discussed by Cecchi-Pestellini & Dalgarno (2000) and Le Petit et al. (2004) for the interpretation of the large $H_2$ column densities measured in diffuse clouds.

Recently it has been theoretically shown (Tsytovich et al. 2014) that interstellar dust is intrinsically unstable against the formation of compact clouds whose size may vary from fractions to dozens of AU. The clouds are formed due to the so-called "shadowing" forces that tend to bind dust together, as illustrated in Figure 1. These forces, exerted between individual grains by surrounding ions and neutrals, represent a generic mechanism of attractive collective interactions in weakly ionized gases (Khrapak et al. 2001). The equilibrium density in a cloud is reached when the shadowing is balanced by the mutual electrostatic repulsion of grains (charged due to a combined effect of photoemission and collection of electrons and ions). It was demonstrated that such clouds should produce gaseous clumps with the local density much larger than the density of ambient ISM.

In this paper we study a possible influence of the compact dusty clouds on the rate of $H \rightarrow H_2$ transition in diffuse ISM. In Section 2 we summarize the mechanisms leading to the formation of dusty clouds and the creation of gaseous clumps around them. In Section 3 we discuss the main processes determining the efficiency of the recombination (association) of atomic hydrogen on dust, and estimate the expected average enhancement of the $H_2$ formation. In Section 4 we mention various mechanisms that have been proposed previously to explain available observational data, indicating the existence of tiny-scale gaseous clumps in diffuse ISM, and argue that the mechanism associated with the compact dusty clouds provides optimum conditions for $H \rightarrow H_2$ transition. Finally, we point out some astrophysical implications of the enhanced $H_2$ formation due to the presence of dusty clouds.

2. DUSTY CLOUDS AND GASEOUS CLUMPS

Compact dusty clouds have been proposed by Tsytovich et al. (2014) to form in diffuse ISM due to at-
Homogeneous dust distribution

Compact dusty clouds

Fig. 2.—A tiny-scale structure of the ISM, associated with compact dusty clouds (not to scale, see Tsytovich et al. 2014). A homogeneous dust distribution is intrinsically unstable against the formation of compact spherical clouds in diffuse ISM, where the surface temperature of dust, $T_{\text{dust}}$, is usually significantly smaller than the gas temperature $T_{\text{gas}}$. The dust density inside the clouds, $n_{\text{dust}}^{(cl)}$, is drastically increased with respect to the density $n_{\text{dust}}^{(h)}$ in a homogeneous case, the resulting average distance between the clouds, $L_{\text{cl}}$, is $\sim \sqrt{n_{\text{dust}}^{(cl)}/n_{\text{dust}}^{(h)}}$ times their size $d_{\text{cl}}$. Efficient radiative cooling ensures that the dust surface temperature is the same in both cases (and equal to $T_{\text{dust}}$). The gas temperature inside the clouds, $T_{\text{gas}}^{(cl)}$, is close to $T_{\text{dust}}$ due to strong thermal coupling; outside the clouds the gas temperature rapidly tends to $T_{\text{gas}}$ (same as in a homogeneous case). Therefore, a gaseous “clump” is formed around each cloud (indicated by the blue shading), where the local gas density is increased by a large factor of $T_{\text{gas}}/T_{\text{dust}}$ with respect to the ambient gas density. While the dust is virtually absent between the clouds, the gas density in this region remains practically unchanged compared to a homogeneous case. Typical values of $d_{\text{cl}}$ in diffuse ISM vary from fractions to dozens of AU.

The shadowing forces compress dust into compact clouds, as sketched in Figure 2, where the equilibrium is reached due to the mutual electrostatic repulsion between charged grains. The dust density inside a cloud, $n_{\text{dust}}^{(cl)}$, can be many orders of magnitude higher than the average dust density $n_{\text{dust}}^{(h)}$ in a homogeneous case: e.g., the maximum value of $n_{\text{dust}}^{(cl)}/n_{\text{dust}}^{(h)}$ for $\sim 0.03$ μm grains is predicted to be as high as $\sim 10^7$ (Tsytovich et al. 2014), this factor only slightly depends on the grain size. Despite such a high local density, the dust coagulation inside the clouds is practically inhibited because of a strong electrostatic repulsion between grains (whose charges $Z$ in diffuse ISM regions are usually positive due to photoemission by the interstellar UV field).\(^1\) Extinction in dusty clouds is governed by the Rayleigh regime, and is very weak in the visible spectral range (Tsytovich et al. 2014). It may be significant in the UV, in a cloud formed by submicron dust (representing the upper-size bound of the MRN-like distributions, e.g., Kim et al. 1994; Weingartner & Draine 2001). However, for small (e.g., $\sim 10^{-7}$ μm) grains, extinction in dusty clouds is negligible.

Dense dusty clouds can, in principle, be distorted by turbulent motion present in the ISM down to sub-AU length scales (e.g., Draine 2011, and references therein). Nevertheless, the generic shadowing mechanisms of dust compression remain unaffected by subsonic velocity perturbations. Since the detected relative fluctuations of the electron density, associated with turbulence at such small scales, are of the order of $10^{-3} - 10^{-4}$ (Cordes et al. 1985), the corresponding velocity perturbations must be very subsonic. Therefore in this paper we assume equilibrium properties of the clouds, as derived by Tsytovich et al. (2014) for a quiescent environment.

2.1. Creation of gaseous clumps

One of the features of dusty clouds, playing – as shown in the next section – the crucial role for the surface chemistry, is an efficient thermal coupling between dust and gas. The coupling is strong because the equilibrium cloud size is self-regulated to be of the order of (or much larger than) the local mean free path of neutrals, which is primarily determined by their collisions with grains (Tsytovich et al. 2014). The right panel of Figure 2 shows the effect on the gas distribution: Due to thermal accommodation of atoms colliding with dust, gas inside the clouds acquires the temperature of the dust surface, $T_{\text{gas}}^{(cl)} \approx T_{\text{dust}}$. As a result, the gas density inside the clouds increases by a factor of $\sim T_{\text{gas}}/T_{\text{dust}}$ with respect to the ambient density,\(^2\) to ensure a constant gas pressure, and thus each dusty cloud produces a local “atmosphere” – a gaseous clump. The conservation of the radial heat flux yields the profiles of the gas temperature

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1 According to Draine (2011), grains larger than $\sim 10^{-2}$ μm are always multiply charged in diffuse ISM; the surface potential $e/Z/a$ of a grain of radius $a$ can be roughly estimated as $\sim 1 - 2$ V (WNM) or $\sim 0.3$ V (CNM). The resulting electrostatic barrier $e^2Z^2/a$ exceeds the thermal kinetic energy $k_BT_{\text{gas}}$ by more than one (WNM) or two (CNM) orders of magnitude.

2 The gas temperature outside the clouds is the same as in the homogeneous case; we denote it for brevity by $T_{\text{gas}}$. 
and density outside the cloud, rapidly approaching the ambient gas values (Tsytovich et al. 2014). We note that the generation of clumps does not practically affect the gas density in the dust-free regions, between the clouds: Taking into account that the average distance between the clouds $L_{cl}$ and their characteristic size $d_{cl}$ are related as $(L_{cl}/d_{cl})^3 \sim n_{dust}^{(b)}/n_{dust}^{(cl)}$, and using the global gas conservation, we obtain that the resulting relative decrease of the gas density (with respect to a homogeneous case) is of the order of $(n_{dust}^{(b)}/n_{dust}^{(cl)})(T_{gas}/T_{dust})$, i.e., is very small for all reasonable values of the temperature contrast.

The creation of clumps occurs at a timescale of the gas diffusion, which is equal to the squared size of a dusty cloud divided by the diffusion coefficient of gas particles. The latter approximately equals to $v_H/\sigma n_H$ (e.g., Smirnov 2006), where $v_H = \sqrt{k_B T_{gas}/m_H}$ is the thermal velocity scale of H atoms, $\sigma \sim 10^{-15}$ cm$^2$ is the gas-kinetic cross section of their mutual collisions, and $n_H$ is their density. Thus, the characteristic time of the clump creation is of the order of $\mathcal{O}(d_{cl}^2/\sigma n_H v_H)$. For typical values of $n_H \sim 10^5$ cm$^{-3}$ and $v_H \sim 10^5$ cm/s this process is very fast, taking a few yr for a 1 AU cloud (Tsytovich et al. 2014). A timescale of the formation of dusty clouds is a factor of $\mathcal{O}(\sqrt{m_{dust}/m_H})$ longer, where $m_{dust}$ is the grain mass and $Z$ is the charge number; e.g., for $\sim 0.03$ µm grains with $m_{dust} \sim 3 \times 10^{-16}$ g and $Z \sim 10$ dusty clouds are formed within $\sim 10^3$ yr. Still, this timescale is much shorter than the average lifetime of a diffuse molecular cloud (over 1 Myr).

It is noteworthy that the photoelectric (and cosmic-ray) heating and radiative cooling, whose balance governs the equilibrium gas temperature in a homogeneous case, cannot noticeably affect the temperature inside the clouds: According to Draine (2011), the gas cooling function $\Lambda$ in diffuse ISM regions rapidly decreases with decreasing $T_{gas}$, and does not exceed a value of $\mathcal{O}(n_H^{-1} \sim 10^{-22})$ erg cm$^{-3}$ s$^{-1}$ for $T_{gas} \sim 10^2$ K (this ratio is practically independent of $n_H$). The resulting characteristic gas cooling time, $n_H k_B T_{gas}/\Lambda \propto n_H^{-1}$, is longer than $\sim 3 \times 10^2$ yr (for $n_H \sim 10^{-3}$ cm$^{-3}$ and $T_{gas} \sim 10^5$ K). This should be compared with the mean time of gas-dust collisions, $\sim (\pi a^2 \Sigma n_{dust}^{(cl)})^{-1}$, which also scales as $\propto n_H^{-1}$. Setting $n_{dust}^{(cl)} \sim 10^3 n_{dust}^{(b)}$ and estimating the characteristic density of ISM grains from the MNR size distribution, $n_{dust}^{(b)}(a) \sim 10^{-25}(a$/cm$)^{-2.5} n_H$ (Weingartner & Draine 2001), we obtain that the collision time for $a \sim 0.03$ µm is 2-3 orders of magnitude shorter than the cooling time. This justifies the assumption of a perfect thermal coupling of gas to dust.

To conclude this section, we point out that the equilibrium cloud size is inversely proportional to the local gas density (Tsytovich et al. 2014). Thus, the creation of a gaseous clump should stimulate the breakup of the original dusty cloud into smaller equilibrium clouds whose characteristic size is a factor of $\sim T_{gas}/T_{dust}$ smaller than the original size, while the local gas density inside these smaller clouds remains unchanged (and equal to $\sim T_{gas}/T_{dust}$ times the ambient gas density). While the breakup kinetics, not considered by Tsytovich et al. (2014), is an interesting problem by itself, it does not qualitatively affect the process of $H_2$ formation, which is the main focus of the present work. Therefore we leave the analysis of this problem for a separate paper.

3. FORMATION OF MOLECULAR HYDROGEN IN DIFFUSE ISM

The presence of dusty clouds makes all chemical reactions, occurring in diffuse ISM at the surface of grains, extremely heterogeneous. Since the rates of the surface reactions are generally nonlinear functions of the gas density, it is natural to expect that also the global rates (averaged over the tiny-scale inhomogeneities) must be affected by the presence of dusty clouds.

Let us elaborate on the effect of dusty clouds on the formation of $H_2$. The equilibrium molecular density in the gas phase is determined by the balance between the photodissociation, which is the principal process destroying interstellar $H_2$, and the atomic recombination at the grain surface (Jura 1974, 1975, Draine 2011, Wakeham et al. 2017). While the photodissociation term in the corresponding balance equation is linearly proportional to the molecular density (the same is true for other relevant dissociation processes, e.g., due to CRs), the recombination term is essentially nonlinear.

In the framework of the basic Langmuir kinetics for the interaction of hydrogen atoms with dust, the equilibrium surface coverage of the physisorbed sites $\varphi$, equal to the areal density of hydrogen atoms multiplied by the site area $S$ (for simplicity we assume $\varphi \ll 1$), is described by the following balance equation (e.g., Bihlm & Lipshtat 2002, Wakeham et al. 2017):

$$S j_H \simeq \varphi v \exp\left(-\frac{E_{des}}{k_B T_{dust}}\right) + 2\varphi^2 v \exp\left(-\frac{E_{diff}}{k_B T_{dust}}\right). \tag{1}$$

The lhs is determined by the effective flux of incoming hydrogen atoms per unit area, $j_H = \pi a^2 \Sigma n_H v_H$ (i.e., $S j_H$ is the effective flux in units of monolayers/s), which is proportional to the sticking probability $s$, a function of the gas temperature. Since typical $T_{dust}$ in diffuse ISM is about 15 K (Tielens 2005, Hocuk et al. 2017), and the sticking probability in chemisorbed sites for local gas temperatures ($\simeq T_{dust}$) is extremely low (Cazaux et al. 2011), the physisorption is assumed to be the main adsorption mechanism. The first depopulation term on the rhs represents the thermally activated desorption of atoms, the second term is due to their recombination induced by thermally activated diffusion, where $E_{des}$ and $E_{diff}$ are the respective activation energy barriers and $v$ is the typical attempt rate characterizing thermal hopping of the adsorbed atoms.

The diffusion energy comprises a certain fraction of the desorption energy, $E_{diff} = \alpha E_{des}$ (Wakeham et al. 2017). The exact value of $\alpha$ is unknown, and for different atoms it may vary from $\sim 0.3$ (Hasegawa et al. 1992) to $\sim 0.7$ (Minissale et al. 2016). The magnitude of $E_{des}$/$k_B$ is estimated to be about 370 K for olivine and about 660 K for amorphous carbon (e.g., Katz et al. 1999). This implies that both depopulation terms on the rhs of Equation (1) have a very sharp dependence on the dust temperature. The desorption and recombination terms dominate the depopulation at higher and lower $T_{dust}$, respectively, and a transition between these two regimes is very sharp, too. Assuming $n_H = 10^2$ cm$^{-3}$ and $T_{gas} = 10^2$ K, which
approximately corresponds to H I regions (Draine 2011), and setting $\nu \sim 10^{12} $ s$^{-1}$, from Equation (1) we infer that the desorption dominates at $T_{\text{dust}} \gtrsim 13$ K for olivine and at $T_{\text{dust}} \gtrsim 23$ K for carbon (for $\alpha = 0.5$). Therefore, in diffuse ISM the surface kinetics of H atoms is expected to occur in the desorption-dominated regime for olivine grains, which represent the main fraction of the interstellar dust (Hocuk et al. 2017). For carbonaceous grains, on the other hand, the recombination term should dominate the kinetics.

The rate of H$_2$ formation per unit dust area, described by the recombination term in Equation (1), scales as $\propto \varphi^2$. In the desorption-dominated regime, where $\varphi \propto j_H$, this yields $R_{\text{H}_2} \propto \varphi^2 j_H n_{\text{dust}}$ for the formation rate per unit volume, where $n_{\text{dust}}$ is the relevant dust density. Note that one can also add the Eley-Rideal (ER) recombination term $\sim \varphi^2 j_H$ (Cazaux & Tielens 2004) to the rhs of Equation (1), which does not change the obtained scaling dependence of $R_{\text{H}_2}$ in this case. In the recombination-dominated regime, where $\varphi^2 \propto j_H$, we obtain $R_{\text{H}_2} \propto j_H n_{\text{dust}}$; the ER term in this case is negligible as long as $\varphi \ll 1$.

3.1. Effect of dusty clouds

The simple consideration above allows us to draw an important general conclusion concerning the role of dusty clouds in the average steady-state abundance of molecular hydrogen. The observationally-relevant (effective) characteristics of the ISM should be derived by averaging over tiny-scale inhomogeneities introduced by the clouds, as $\langle \ldots \rangle = \lambda^{-1} \int \lambda \ldots d\lambda$, where $\lambda \sim L^3_{\text{cl}}$ is the ISM volume per dusty cloud (see right panel of Figure 2).

We point out that the characteristic timescale of H$_2$ formation in diffuse ISM is significantly longer than the diffusion time of hydrogen at the intercloud distance $L_{\text{cl}}$. For typical parameters $L_{\text{cl}} \sim 10^2 d_{\text{cl}} \sim 10^2$ AU, $n_H \sim 10^2$ cm$^{-3}$, and $\sigma \sim 10^{-15}$ cm$^2$, the diffusion time is $\sim L_{\text{cl}}^2 \sigma n_H / v_H \sim 3 \times 10^5$ yr, which is about two orders of magnitudes shorter than the H$_2$ formation timescale for the same density (Draine 2011). Therefore, it is reasonable to assume that the molecular hydrogen generated in the clumps is homogeneously distributed across the ISM volume due to diffusion.

In a H$_2$ balance equation, the (photo)dissociation term scales as $\propto n_{\text{H}_2}$, and thus is obviously not affected by the averaging. On the other hand, the integral over the formation rate $R_{\text{H}_2} \propto \varphi^2 j_H n_{\text{dust}}$ is non-zero only within the cloud volume, where the dust density is $n_{\text{dust}} (cl)$ and $\beta$ varies between 1 (desorption-dominated regime, higher $T_{\text{dust}}$) and 1/2 (recombination-dominated regime, lower $T_{\text{dust}}$). The value of $j_H$ is proportional to the product of the local density of hydrogen atoms and the square root of the local temperature. Since the gas pressure is constant across a clump, we have $j_H \propto \sqrt{n_H}$. Setting the local gas density $n_H$ inside dusty clouds equal to the ambient gas density multiplied by a factor of $\sim T_{\text{gas}} / T_{\text{dust}}$, and utilizing the global dust conservation, we derive the effective rate of H$_2$ formation,

$$ (R_{\text{H}_2}) \simeq \left( \frac{T_{\text{gas}}}{T_{\text{dust}}} \right)^{\beta} R_{\text{H}_2}, $$

where $R_{\text{H}_2}$ is the formation rate corresponding to a homogeneous case (left panel of Figure 3).

Thus, the relative enhancement of the global formation rate in the presence of dusty clouds (and, hence, the increase of the molecular abundance in diffuse H I regions) is expected to be about $(T_{\text{gas}} / T_{\text{dust}})^2$. For a typical dust temperature about 15 K and gas temperatures varying between 80 K and 150 K toward different lines of sight (e.g., Falgarone et al. 2005, Snow & McCaill 2006, Gerin et al. 2015, Winkel et al. 2017), one can expect that H$_2$ formation will be accelerated by a factor of 5–10.

4. DISCUSSION AND CONCLUSION

There is a great deal of observational evidence indicating strong inhomogeneity of diffuse ISM at the length scales down to $\sim$ AU (e.g., Dieter et al. 1976, Crawford 2002, Lazio et al. 2009, Stanimirovic et al. 2010, Dutta et al. 2014, Corby et al. 2018). Various mechanisms proposed so far to explain this phenomenon include the formation of cold anisotropic (sheetlike) structures (Heiles 1997, 2007), fractal structures arising from MHD turbulence (Emsenmacher 1999) and irregularities representing the tail-end of the turbulent spectrum (Deshpande 2000), structures generated due to excitation of slow magnetosonic waves (Falke & Hartquist 2002, Hartquist et al. 2003), etc. However, only the mechanism associated with the formation of compact dusty clouds enables creation of equilibrium gaseous clumps. This latter feature is particularly important for the H$_2$ formation, providing optimum conditions for the heterogeneous enhancement of the atomic recombination discussed in Section 3.1.

The results of Section 3.1 show that the existence of gaseous clumps around dusty clouds in diffuse ISM may shorten the timescale of H$\rightarrow$H$_2$ transition by up to an order of magnitude, thus reducing it to a few Myr in our model. The timescale of H$_2$ formation has a broad astrophysical importance – in particular, it determines the possible physical scenarios of evolution of giant molecular clouds (GMCs). It has been suggested that long timescales (exceeding $10^7$ yr) favor the scenario of slow evolution of typical GMCs (with $n_H \sim 10^2$ cm$^{-3}$, Blitz & Shu 1980) as gravitationally bound objects in virial equilibrium, supported either by magnetic fields or by turbulence (Tassis&Mouschovias 2004, Mouschovias et al. 2006, Matzner 2002, Krumholz et al. 2006).

An alternative theory of formation of GMCs implies that they are the transient objects in diffuse medium, formed and destroyed by large-scale turbulent flows on timescales of $\sim$ Myr (e.g., Mac Low & Klessen 2004). This is inconsistent with the timescales of H$_2$ formation in a homogeneous quiescent gas (Hollenbach et al. 1971). Furthermore, there are indications that the lifetime of molecular clouds may indeed be relatively short – these include, e.g., the lack of post-T Tauri stars older than 3 Myr in nearby star-forming regions and the absence of mechanisms that can prevent fast gravitational collapse and fragmentation of newly-formed clouds (Hart-
mannon et al. 2001).\(^4\) Also, the high fraction of molecular hydrogen observed in turbulent diffuse ISM implies \(\text{H}_2\) formation timescales on the order of \(10^6\) yr (see Liszt 2007, and references therein).

To the best of our knowledge, all existing scenarios of rapid \(\text{H} \rightarrow \text{H}_2\) transition in diffuse ISM and formation of GMCs assume inhomogeneities in gas and dust densities caused by dynamical processes, such as cloud collisions or turbulence on different scales (e.g., Glover & Mac Low 2007; Lesaffre et al. 2007; Godard et al. 2009; Micic et al. 2012; Valdivia et al. 2016). The possible existence of equilibrium gaseous clumps, considered in this work, leads to a new type of physical mechanism that can significantly accelerate the \(\text{H}_2\) formation in diffuse medium and foster the efficient \(\text{H} \rightarrow \text{H}_2\) transition. Obviously, chemistry of other species in diffuse ISM is also affected by possible presence of such clumps, but we leave this question for a separate study.

\(^4\) Note, however, that a rapid star formation scenario proposed by Hartmann et al. (2001) for the Taurus-Auriga association is debated by Palla & Stahler (2002).

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