Depolarization of Spin-Polarized hydrogen via Spin-Exchange Collisions with chlorine Atoms at Ultrahigh Density

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Recently, the production of ultrahigh-density (> 10^{19} \text{cm}^{-3}) spin-polarized D atoms was demonstrated, from the photodissociation of deuterium iodide, but the upper density limit was not determined. Here, we present studies of spin-polarized hydrogen (SPH) densities up to 10^{20} \text{cm}^{-3}, by photodissociating 5 bar of hydrogen chloride with a focused 213 nm, 150 ps laser pulse. We extract the depolarization cross-section of hydrogen and chlorine atom collisions, which is the main depolarization mechanism at this high-density regime, to be $\sigma_{\text{H-Cl}} = 7(2) \times 10^{-17} \text{cm}^2$. We discuss the conditions under which SPH densities of 10^{20} \text{cm}^{-3} can be reached, and the potential applications to ultrafast magnetometry, laser-ion acceleration, and tests of polarized nuclear fusion.

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I. INTRODUCTION

Polarized atomic gases typically have low densities, of $\sim 10^{12} \text{cm}^{-3}$ and below, because of depolarizing effects of collisions in the production methods of Stern-Gerlach spin separation [1], or optical pumping [2, 3]. The only exceptions have been the noble gases with nuclear spin (e.g. $^3\text{He}$ and $^{129}\text{Xe}$), for which the nuclear spins can be polarized at high pressure (via spin-exchange optical pumping), as the depolarization rate of these closed-shell atoms can be very small [4]. However, this inability to produce high-density open-shell spin-polarized gases precludes several potential applications.

Recently, Sofikitis et al. demonstrated the production of high-density ($\sim 10^{19} \text{cm}^{-3}$), highly spin-polarized hydrogen (SPH) and deuterium (SPD), from the photodissociation of hydrogen halides with a circularly polarized UV laser pulse, and a pickup coil to monitor the electron polarization [5]. These densities surpass the current state-of-the-art by at least 7 orders of magnitude. To achieve such high densities, the hydrogen halides were photodissociated with a 150 ps laser pulse, so that the SPH/SPD are produced nearly instantaneously, on the 10^{-10}s timescale, several orders of magnitude faster than the production time for conventional polarization methods. This rapid production allows the high-density SPH/SPD to survive for 5-100 ns at these high densities [6], before depolarization from collisions. The aim of this paper is to investigate the main depolarization mechanism in detail, and to determine the limits of SPH density and lifetime.

The main SPH depolarization mechanism at ultrahigh densities is expected to be spin-exchange collisions between SPH and halogen radicals Y, occurring at a rate $k_\text{H-Y}$:

$$H^\uparrow + Y \rightarrow H + Y^\uparrow$$

as opposed to the low-density regime ($\sim 10^{16-17} \text{cm}^{-3}$), where only $\sim 0.1\% \text{HCl}$ is dissociated, and the prevailing depolarization mechanism has been shown to be the three-step reaction of SPH with high-density HY molecules [6]:

$$H^\uparrow + HY \rightarrow HY - H^\uparrow \quad (1)$$

$$HY - H^\uparrow \rightarrow HY - H \quad (2)$$

$$HY - H^\uparrow + HY \rightarrow H^\uparrow + 2HY \quad (3)$$

To our knowledge, the depolarization cross-section of SPH by halogen radicals is not cited in the literature. If we assume that $\sigma_{\text{H-Y}}$ is comparable to the depolarization cross-section of SPH by alkali atoms, $\sigma_{\text{H-Rb}} \sim 2 \times 10^{-15}$ cm$^2$, then, for a density of $10^{19} \text{cm}^{-3}$, the polarization lifetime would be $\sim 0.2$ ns, which is faster than a hyperfine beating cycle for both H and D. Sofikitis et al. [5] used a simple method to estimate $\sigma_{\text{H-Y}}$. They focused the beam lenses, one with $f=25$ mm and one with $f=50$ mm, placed at a distance $l \sim 25 \text{mm}$ from the center of the coil. The $f=50$ mm lens focused the beam $\sim2.5$ cm away from the center of the coil, and produced an SPH density of $\sim 10^{16} \text{cm}^{-3}$ inside the coil, whereas the $f=25$ mm lens focused the beam at the center of the coil, creating an
effective SPH density of $\sim 10^{19}$ cm$^{-3}$ near the focus. This way, they expected to differentiate between the low- and high-density depolarization rates. However, they observed lifetimes of $\sim 10 - 20$ ns under both density regimes, yielding no evidence of SPD depolarization from $I(2^2P_{3/2})$ in the high-density regime. The deuterium iodide (DI) density could not be increased above $\sim 10^{19}$ cm$^{-3}$, to increase the depolarization rate, because the absorption cross section of DI at 266 nm, $2 \times 10^{-19}$ cm$^2$, is large enough to prevent sufficient photolysis laser light reaching the laser focus, for the geometry of the experiments. The absorption cross section of HCl at 213 nm is 2 orders of magnitude lower, at $2 \times 10^{-21}$ cm$^2$. Therefore, the HCl density can be increased to $10^{20}$ cm$^{-3}$, and the photolysis laser can reach the laser focus in the cell, without too much loss from absorption, as the optical depth is $\sim 5$ cm. Therefore, we are able to study the SPH depolarization dynamics at significantly higher densities, needed to determine the SPH-Cl depolarization cross section. In the following sections, we describe the experimental method and analysis to determine $\sigma_{H - Cl}$.

II. HALOGEN RECOMBINATION

The time dependent halogen-atom density is determined by the recombination rate, so this recombination is first discussed in detail in this section. The recombination reactions are three-body reactions, of the form:

$$2Y + M \rightarrow Y_2 + M$$ (4)

Depending on which third body M participates in the reaction, the reaction rate is different. As shown in table II, the three-body Cl recombination reaction rates are of order $10^{-32}$ cm$^6$ s$^{-1}$ at room temperature, when the third body is a chlorine molecule, and somewhat lower in the presence of $N_2$. Nonetheless, immediately after the photodissociation, no halogen molecules are present; recombination reactions with $M= Y$ or $M= HY$ should take place to create $Y_2$, to make the three-body recombination reactions faster. Reactions with $M= Y$ are significantly slower than those with $M= Y_2$, having a rate of $3.5 \times 10^{-33} e^{810/T}$ cm$^6$ s$^{-1}$ [8]. Furthermore, to our knowledge, there are no references of the rate of recombination reactions with a HY third-body. These rate constants do not justify the existence of a significant $Y_2$ density immediately after the photodissociation, therefore, we can assume that the Y atom density remains constant for the few ns after photodissociation that are of interest to this study.

A number of inert gases, such as $N_2$, $SF_6$, or $C_2F_6$ are efficient third bodies for halogen recombination. For example, recombination of Cl radicals in the presence of $SF_6$ molecules happens at a rate $k_{Cl - SF_6} = 6.6 \times 10^{-33}$ cm$^6$ s$^{-1}$ [9].
This reaction rate is enough to create a population of $\text{Cl}_2$ comparable to the radical population in a few tens of ns at pressures of 2 bar each of HCl and $\text{SF}_6$ (1d). Such a high $\text{Cl}_2$ concentration, though, would trigger the efficient reaction:

$$\text{H}^\uparrow + \text{Cl}_2 \rightarrow \text{HCl} + \text{Cl}$$

which removes SPH, while also creating Cl radicals, an effective SPH depolarizer. Thus, the fast recombination that removes Cl, triggers another reaction that replenishes it and simultaneously depolarizes SPH. Taking these reactions into account, the rate equations for the system after the UV pulse photodissociation are:

$$\frac{d[\text{SPH}]}{dt} = -k_{\text{H}-Y}[\text{SPH}][Y] - k_{\text{H}-\text{HY}}[\text{SPH}][\text{HY}] - k_{\text{H}-\text{Y}_2}[\text{SPH}]$$

$$\frac{d[Y]}{dt} = k_{\text{H}-\text{Y}_2}[\text{SPH}][Y]$$

$$\frac{d[Y_2]}{dt} = k_{\text{Y}-\text{Y}-M}[Y]^2[M] - k_{\text{H}-\text{Y}_2}[\text{SPH}][Y]$$

$$\frac{d[\text{HY}]}{dt} = k_{\text{H}-\text{Y}_2}[\text{SPH}][Y]$$

The rate constants for 5 are shown in table I. All are of order $10^{-11} \text{cm}^3 \text{s}^{-1}$, implying that, for $Y_2$ densities of $10^{19} \text{cm}^{-3}$ (similar to the corresponding SPH densities after photodissociation), SPH is removed in less than a ns. We can therefore safely conclude that, if the conditions of the experiment favored a very rapid recombination of the Y radicals, a short SPH lifetime would reflect this.

Concluding this section, we can say that the observed SPH and SPD lifetimes in [5] are shorter than the limits imposed by the halide recombination dynamics described above. Therefore, any depolarization observed should be attributed to spin-exchange reactions of SPH with the Y radical.

| $\text{Y}_2$ | $Y_2$ |
|---|---|
| H + Cl$_2$ | $2.13 \times 10^{-11}$ |
| D + Cl$_2$ | $1.4 \times 10^{-11}$ |
| H + Br$_2$ | $8.2 \times 10^{-11}$ |
| D + Br$_2$ | $5.6 \times 10^{-11}$ |

TABLE I: Rates of the reaction $H + Y_2 \rightarrow HY + Y$ in (cm$^3$ s$^{-1}$)[10]

| Y | M | $Y_2$ | Ar | $N_2$ | Pressure (bar) |
|---|---|---|---|---|---|
| Cl | $5.7 \times 10^{-32}$ | $5.5 \times 10^{-32}$ | $1.13 \times 10^{-32}$ | $1.38 \times 10^{-32}$ | $180$ [11] |
| | $5.8 \times 10^{-32}$ | $1.2 \times 10^{-32}$ | $10^{-32}$ | $5.33 \times 10^{-3}$ [13] |
| Br | $1.22 \times 10^{-32}$ | $4.25 \times 10^{-33}$ | $6.07 \times 10^{-33}$ | $9.1 \times 10^{-33}$ | $100$ [14] |
| | $3.01 \times 10^{-33}$ | $6 \times 10^{-33}$ | $9 \times 10^{-33}$ | $4.27 \times 10^{-3}$ [15] |

TABLE II: halogen three-body recombination rates for various chaperons M in cm$^6$ s$^{-1}$

III. DETERMINATION OF $\sigma_{H-Y}$

The rate equations for SPH depolarization from collisions with Y radicals read as
FIG. 2: Plot of the spin polarized Hydrogen density inside the coil, after the photodissociation of 2 bar HCl. The lower range of the plot densities is 0.1%

\[
\frac{d[SPH]}{dt} = -k_{H-Y}[SPH][Y] - k_{H-HY}[SPH][HY] \tag{10}
\]
\[
\frac{d[Y]}{dt} = 0 \tag{11}
\]
\[
\frac{d[HY]}{dt} = 0 \tag{12}
\]

where we neglect all \( Y_2 \) recombination reactions, since they are significantly slower than the depolarization lifetime. It follows that, to calculate \( k_{H-Y} \), the SPH (and \( Y \)) density immediately after the photodissociation must be known. If the photodissociation is 100% efficient, \( HY(0) = 0 \), and the second term of 10 vanishes. Otherwise, the second term is non-negligible, and can be calculated using the value of \( k_{H-HY} \) from [6] \( (7.5 \times 10^{-19} \text{cm}^2) \).

Note that, as mentioned in [5], and was confirmed in this study, no evidence of spin-polarized halogens (i.e., a Fourier peak at 150 MHz, the hyperfine frequency of Cl \( ^2P_{3/2} \)) is found in the data. The absence of such evidence means that halogen atoms are quickly \((\ll 1 \text{ ns})\) depolarized. This supports the conclusion that the main depolarizer of SPH is the unpolarized halogen atom; if the halogen atom retained its polarization for a longer time, the H–Y spin-exchange cross section would be much lower.

A. Experiment

A simple experiment was designed to estimate \( k_{H-Y} \), by measuring the SPH polarization lifetime for various SPH densities. The experimental setup is the same as the one used in [5], with hydrogen chloride (HCl) as the SPH source gas. Briefly, 1-5 bar HCl are introduced in a gas cell, and a circularly polarized, 213 nm, 150 ps laser pulse is used to photodissociate HCl. The 4 mm diameter beam is focused by a 5 cm lens, placed on a one-dimensional translation stage, by which we control the distance of the lens from the cell, and therefore the position of the focus inside the cell. A pickup coil with a length of 4 mm, 4.5 turns, and a 2 mm diameter, is used to detect the time evolution of the SPH produced by the photodissociation of HCl. The time-dependent signal that the coil picks up is of the form:

\[
I(t) = e^{-t/\tau} \cos 2\pi ft \tag{13}
\]

Where \( t \) is the time, \( \tau \) the polarization lifetime, and \( f \) is the hyperfine beating frequency, which is equal to 1.420415 GHz for the unperturbed hyperfine hydrogen Hamiltonian

\[
H = A_{HF} \mathbf{I} \cdot \mathbf{J} \tag{14}
\]

with \( A_{HF} \) the hyperfine constant, and \( \mathbf{I}, \mathbf{J} \) the quantum operators for the nuclear and electron spin, respectively. With this setup, we can scan the position of the focus through the coil length, effectively varying the density of the SPH produced inside the coil.
The dimension of the beam was confirmed to follow the distribution of a Gaussian beam using a razor-edge scheme. The waist at focus was estimated to be $6 \pm 1 \mu m$, close to the diffraction limit of $d = 5 \mu m$. A theoretical model based on the saturated Beer-Lambert law is used to simulate the densities created by a laser beam of known dimensions and energy. The produced SPH density through the length pickup coil length, is shown in figure 2, for an HCl pressure $P=2$ bar, corresponding to an initial number density of $5.4 \times 10^{19} cm^{-3}$. The model shows that the SPH density within the Rayleigh range of the beam ($l \sim 0.5 mm$) is close to the initial HCl density for pressures of up to 5 bar. However, due to the large beam divergence and the low absorption cross-section of hydrogen chloride, the density outside of the Rayleigh range is much lower, of order $10^{17-18} cm^{-3}$. If SPH depolarization at a high density environment is fast, this variation in density within a few mm would result in polarization lifetime gradients in space. Since the pickup coil is longer than this length, the different SPH polarization lifetimes would be imprinted in the pickup coil signal (13) as more than one exponential:

$$I(t) = \int \int \rho_{SPH}(r, \theta) e^{-t/\tau(r, \theta)} \cos(2\pi ft) \, dr \, d\theta$$

where $\rho_{SPH}$ and $\tau$ depend on $r$ and $\theta$.

A shorter coil would, of course, offer better spatial resolution, but at the detriment of the signal-to-noise ratio (SNR). For this reason, in this experiment, we chose to work with a 4.5 turns, 4 mm long pickup coil. Further investigation on the optimization of the coil is needed, to achieve shorter length while retaining a high SNR.

**B. Calculation of the Expected SPH Density**

The laser used in this experiment emits a $\lambda = 213$ nm, $\tau_p = 150$ ps, $E = 3$ mJ, $D = 4$ mm Gaussian pulse. The pick-up coil’s diameter and length are $d = 2$ mm and $l = 3.5$ mm respectively, and is placed 3 cm away from the window of the cell. The coil is relatively short in order to facilitate the acquisition of rapidly changing signal. The photodissociation cross section for HCl at 213 nm (room temperature) is $\sigma = 1.7 \times 10^{-21} cm$[18]. The number of SPH fragments created can be calculated as follows. The initial available photons are:

$$N_{ph0} = \frac{\lambda E}{hc}$$

with $h$ is Planck’s constant, and $c$ the speed of light. Applying the saturated Beer-Lambert law, we find that the photons that reach the coil are:

$$N_{ph} = N_{ph0} e^{-\sigma ln(1 - e^{-\sigma ln})}$$

where $n$ is the particle density and is related to the pressure $P$ (in bar) for HCl as:

$$n = P \times 2.7 \times 10^{19} cm^{-3}$$

**IV. RESULTS AND DISCUSSION**

From figure 2, we can observe the variation in SPH density inside the coil. Inside the Rayleigh range of the beam, HCl is almost or entirely depleted (70-100%), and there exist only hot spin-polarized hydrogens and chlorines immediately after the photodissociation. As discussed earlier, Cl is almost entirely depolarized within less than 1 ns after the photodissociation, thus making SPH-Cl spin-exchange more efficient. The fast depolarization would then be triggered by spin-exchange collisions between SPH and unpolarized Cl. At positions outside the Rayleigh range, SPH and Cl account for less than 1% of the total number density, and are surrounded by HCl molecules. In that case, the main depolarization mechanism is via the HCl-SPH complex formation (1), and the depolarization rate is more than an order of magnitude lower. Using this density calculation for H, Cl, and HCl at every position of the beam, we can extract a value for the H-Cl depolarization cross section from eqs. 10. To that end, a Finite Element Analysis (FEA) is employed, where the volume of the beam is separated in sections of $\{x, y, z\}=1 \mu m \times 1\mu m \times 20\mu m$, and the time evolution of the SPH in each section is calculated.
To extract $\sigma_{\text{H} - \text{Cl}}$, the distribution of velocities should also be calculated. The velocity of SPH after HY photodissociation is $\tilde{v} \approx 20 \text{ km/s}$\[19\]. However, thermal equilibrium between H, Cl (and HCl, where less than 100% of HCl molecules have been photodissociated) should be reached quickly after the photodissociation, as the hard-sphere collision rate is 40/ns.

Using the Equipartition theorem, the kinetic energy of the H atoms, $E_H$, as a function of the dissociated fraction $x$ of the HCl molecules, is given by:

$$E_H = E_{H_{300K}} + E_{EKE} \times \left[ \frac{3x/2}{3x + \frac{C_{\text{HCl}}}{R} (1 - x)} \right]$$

(19)

where $E_{H_{300K}} = 0.038 \text{ eV}$ is the thermal energy of hydrogen at room temperature, $E_{EKE} = E_{\text{photon}} - E_{\text{bond}} = 1.4eV$ is the excess kinetic energy from the photodissociation, $C_{\text{HCl}}$ the heat capacity of HCl, and $x$ the fraction of photodissociated molecules, $x = n/N_0$, with $n$ the SPH density produced by the photodissociation, and $N_0$ the number density of HCl prior to the photodissociation.

Figure 3 shows the equilibrium SPH velocity as a function of the SPH density. We see that the thermal equilibrium velocity of hydrogen when HCl is completely photodissociated is reduced to $\sim 12 \text{ km/s}$, as a result of the frequent collisions with the initially much colder($\sim 300 \text{ K}$) Cl atoms, whereas at low SPH densities, SPH velocity is the room temperature velocity, $\sim 2.5 \text{ km/s}$, given by the room temperature HCl.

We can now use the H, Cl, and HCl density distribution, the SPH-Cl and SPH-HCl relative velocities distribution in eq. 10 to determine $\sigma_{\text{H} - \text{Cl}}$. Figure 5 show the obtained SPH signal after photodissociation by the laser pulse, and a fit with $\sigma_{\text{H} - \text{Cl}} = 7 \times 10^{-17} \text{ cm}^2$, for various focusing geometries, and at two initial HCl pressures, 2 and 5 bar.

When the beam is focused inside the coil, there is a steep drop observed in the early part of the signal, followed by a lower slope at later times. The steep drop should be attributed to the high density inside the Rayleigh range, where the main depolarization mechanism is via spin-exchanging collisions between SPH and Cl. At later times, the slope becomes gradually lower, approaching the low-density depolarization rate. The value of $\sigma_{\text{H} - \text{Cl}} = 7 \times 10^{-17} \text{ cm}^2$ fits the data well at all focusing geometries and at both 2 bar and 5 bar, lending confidence to the determination of the SPH-Cl spin-exchange cross section.

Note that an inert gas with a high heat capacity can be used, to cool the spin-polarized hydrogen down to a lower equilibrium temperature, and as such lower the collision rate, and thus the spin-exchange collision rate(assuming that the spin-exchange cross section does not increase too much at low energies).

A suitable candidate may be hexafluoroethane ($C_2F_6$), which has a heat capacity of 106 J mol$^{-1}$K$^{-1}$ at room temperature, which raises to over 170 J mol$^{-1}$K$^{-1}$ at high temperatures. Additionally, $C_2F_6$ is transparent at middle
and far ultraviolet, reducing the possibility of reacting fragments emerging from dissociation or ionization. By adding an extra factor $k E_{C_2F_6}$, where $k = N_{0C_2F_6}/N_{0HCl}$ the number density ratio of $C_2F_6: HCl$, and $E_{C_2F_6}$ the equilibrium thermal energy of $C_2F_6$, eq. 19 becomes:

$$E_{H_a} = E_{H_{300K}} + E_{KE} \times \frac{3x/2}{3x + \frac{C_{HCl}}{C_{H}} (1 - x) + k \frac{C_{PCF_6}}{C_{PH}}}$$

(20)

The equilibrium velocity of SPH after HCl photodissociation in the presence of various partial pressures of $C_2F_6$ is shown in figure 3. The reduction of the equilibrium velocity by a factor of 3 observed with a 3:1 $C_2F_6/HCl$ ratio is expected to prolong the polarization lifetime by an equal factor at the high-density regime.

While the degree of electronic polarization of SPH can be practically 100%, as polarization is instantaneous, the polarization of the nucleus occurs after a hyperfine half-period, which is $\sim 350$ ps for SPH and $\sim 1.52$ ns for SPD. Therefore, the degree of polarization of the SPH nucleus depends on the polarization lifetime. In fig. 4, we see a simulation of the evolution of the polarized SPH density, after complete photodissociation of 2 bar HCl with a 213 nm laser pulse, in the presence of 6 bar $C_2F_6$. The degree of nuclear polarization that can be achieved with this method is 60%, at an initial SPH density of $5.4 \times 10^{19}$ cm$^{-3}$.

V. CONCLUSIONS

To our knowledge, the value of $\sigma_{H-Cl}$ presented here is the first experimentally measured value of the H-Cl spin-exchange rate, and is consistent with the upper limit of $\sigma_{D-I}$ that Sofikitis et al.[5] suggested in an experiment conducted at one order of magnitude lower densities for DI photodissociation. The measured value shows that lifetimes of order of a few ns are possible, for SPH at densities close to $10^{20}$ cm$^{-3}$. Calculation of $\sigma_{H-Cl}$, particularly as a function of collision energy, will be helpful to corroborate the results and conclusions of this work.

The experimental method presented here could be improved to achieve the capability to test higher SPH densities with a better spatial resolution. For better spatial resolution, a shorter coil is required, which, however, would reduce the inductance and quality factor, and consequently the signal-to-noise ratio. A potential solution for this would be the use of a microstrip coil, which offers a high quality factor at GHz frequencies at a very compact size. Furthermore, higher energy lasers would produce higher SPH densities, possibly without the need for tight focusing, thus eliminating the SPH distribution gradients that the 3 mJ laser used in this study creates. Ultrahigh density SPH has several novel applications, including ns-resolved magnetometry, laser-ion acceleration, and tests of polarized nuclear fusion [20]. Using SPH for magnetometry allows improvement in time resolution of atomic magnetometers by several orders of magnitude, and with spatial resolution of about 10 $\mu$m [21]. Laser-ion acceleration can produce intense, GeV-scale,
electron or proton pulses, from gas targets of density $\sim 10^{19}\text{cm}^{-3}$, which have been unpolarized; only SPH production of HY photodissociation has demonstrated polarized electron, proton, and deuteron densities of $\sim 10^{19}\text{cm}^{-3}$, sufficient for laser-ion acceleration [22–25]. It is known that polarized nuclei increase the fusion cross sections of the D+T and D + $^3He$ reactions by $\sim 50\%$, whereas the effect for the D + D reaction is not known. DI photodissociation can offer sufficiently dense SPD for tests of polarized laser fusion for these three reactions [5, 20, 26]. The signal for the D + D reaction is quadratic in the SPD density (whereas the dependence is linear for the other two reactions), therefore the SPH/SPD density limits determined in this paper are important for calculation of potential reaction yields.
FIG. 5: Experimental Data (Blue) and fits generated by finite element analysis of the produced SPH inside the pickup coil (Light Red) for different positions of the beam focus related to the coil center, at an initial HCl pressure of 2 bar (upper panel) and 5 bar (lower panel). The value for $\sigma_{H-C}$ used is $7 \times 10^{-16}$ cm$^2$. $x$ is the distance of the lens relative to the position where the beam is focused at the center of the coil.
FIG. 6: Same as fig. 5, with the beam focused inside the coil, and two different values for the H-Cl cross section, $\sigma_{\text{H-Cl SE}} = 10^{-17}$ cm$^2$ (left) and $\sigma_{\text{H-Cl SE}} = 5 \times 10^{-16}$ cm$^2$ (right)
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