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Adsorbate dynamics on a silica-coated gold surface measured by Rydberg Stark spectroscopy

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Abstract
Trapping a Rydberg atom close to a surface is an important step towards the realisation of many proposals for quantum information processing or hybrid quantum systems. One of the challenges in these experiments is posed by the electric field emanating from contaminations on the surface. Here we report on measurements of an electric field created by $^{87}$Rb atoms adsorbed on a 25 nm thick layer of SiO$_2$, covering a 90 nm layer of Au. The electric field is measured using a two-photon transition to the $D_2^{3/2}$ and $S_2^{5/2}$ states. The electric field value that we measure is higher than typical values measured above metal surfaces, but is consistent with a recent measurement above a SiO$_2$ surface. In addition, we measure the temporal behaviour of the field and observe that we can reduce it in a single experimental cycle, using ultraviolet light or by mildly locally heating the surface with one of the excitation lasers, whereas the buildup of the field takes thousands of cycles. We explain these results by a change in the adatom distribution on the surface. These results indicate that, while the stray electric field can be reduced, achieving field-free conditions above a silica-coated gold chip remains challenging.

Keywords: atom chip, Rydberg atoms, cold atoms, adsorbates electric fields

1. Introduction
The investigation of Rydberg atoms close to a surface is of great importance and interest for areas ranging from surface physics [1–3] to quantum information processing [4–8], particularly in the context of atom chip [9, 10] experiments. The strong interaction between Rydberg atoms over large distances [11] makes them natural candidates for efficient entanglement mechanisms in cold atom physics [12–15]. They can also help to investigate the transport of excitation energy [16]. In addition, their high sensitivity to environmental influences [11] makes Rydberg atoms suitable as a surface probe.

However, noise sources can strongly influence Rydberg atoms’ energy levels. A typical example of a noise source in atom chip experiments is stray electric fields caused by ad-atoms that stick to the surface during the experimental cycle [17, 18]. The interaction between the adatoms and the surface leads to effective electric dipoles on the surface, building up to a macroscopic electric field in the proximity of the surface [19, 20]. Atom chip experiments have to address this issue, and several methods to reduce the surface fields have been suggested [6, 18, 21].
Here we report on the measurement of electric stray fields emanating from a surface, which consists of a 90 nm Au layer covered with 25 nm of SiO₂, see figure 1(a). We measure the Stark shift, induced by DC electric fields, of the 23D_{5/2} and 25S_{1/2} states of ⁸⁷Rb and retrieve field gradients perpendicular and parallel to the atom chip. From our measurements we infer that the Rb adatoms are the main source for the stray electric fields. Then we examine several methods for reducing the stray fields. We observe the influence of ultra-violet (UV) light at 365 nm, provided by an array of light emitting diodes (LEDs), and of the 480 nm excitation laser on the stray fields. Lastly, we put a special emphasis on the temporal dynamics of both effects.

2. Methods

The experiments are performed in an atom chip setup that features a micro-structured FePt layer for the creation of magnetic micro-trap potentials [23, 24]. This layer is covered with an Au and a SiO₂ coating, which acts as a mirror for a mirror magneto-optical trap (MOT) configuration [25]. Beneath the Au there is an additional 1 μm thick layer of SU8 polymer. A schematic of the apparatus is shown in figure 1(a). The atoms are collected in the MOT from a background vapor of ⁸⁷Rb. They are then optically pumped into the [F, m_f] = [2, 2] Zeeman sublevel, and transferred into a magnetic Ioffe–Pritchard (IP) trap [26] formed by a z-shaped wire located behind the Si substrate (not shown in figure 1). We cool the cloud by forced RF evaporation to ~30 μK and position it at different distances of 10–200 μm below the atom chip by varying the bias magnetic field.

The cloud is then exposed for 100 μs to two laser beams with wavelengths 480 and 780 nm both having ~100 μm beam waist (1/e² radius), which is smaller than the axial size of the cloud but comparable to the radial size. We use a two-photon process with a detuning of Δ/2π = +1.5 GHz from the intermediate 5P_{3/2} level, see figure 1(b) for a level diagram. The laser powers are 140 mW and 80 μW respectively. The beams propagate in the z-direction (perpendicular to the atom chip surface) with linear polarisation, such that both contain σ⁺ and σ⁻ polarisation with respect to the magnetic field at the trap minimum B\text{IP}, which is tilted by a few degrees from the x-direction. Both beams are reflected back by the Au surface, such that they overlap with the incoming beam. The two-photon transition excites the exposed atoms to either the 23D_{5/2} or 25S_{1/2} state, from which they decay to non-detectable states or are ionised, such that they appear as lost from the absorption image. The image is taken shortly (100 μs) after the excitation pulse to prevent neighbouring losses. The ratio of the number of atoms in the “hole” area (N_hole) to a “reference” area (N_ref), both marked in the cloud image in (c), is plotted as a function of the frequency of the 480 nm laser. Note that this ratio does not reach 1 because N_hole < N_{ref}, even without excitation lasers. The detuning is relative to the transition frequency as measured by electromagnetically induced transparency measurements in a room temperature vapour cell without electric and magnetic fields. This zero-detuning frequency is kept throughout this paper. The spacing between the different transitions is 7.4 ± 0.3 MHz, corresponding to B\text{IP} = 4.4 ± 0.2 G at the trap bottom. From independent measurements of the trap bottom we get B\text{IP} = 4.2 ± 0.1 G.
atoms from refilling the depleted area. In addition, we verify that exposing the atoms to the 480 nm or 780 nm light separately did not lead to visible depletion. See figure 1(c) for a sample optical density image of the cloud immediately after the excitation pulse showing the lost atoms as a hole, and the tilt between $B_{HP}$ and the $x$-direction. The typical duration of one experimental cycle is $\sim 20s$.

The number of atoms in the depleted area is normalised against an unexposed reference area in the cloud. This greatly suppresses noise from shot-to-shot fluctuations of the overall number of atoms and improves the visibility of the spectrum. Both lasers are frequency stabilised to a high-finesse cavity using a sideband-locking scheme with typical linewidths of less than 10 kHz [27]. We can either the frequency of the 480 nm laser by changing the sideband frequency of the locking scheme, or the frequency of the 780 nm light by an acousto-optical modulator. The first method is used in figure 5, whereas the latter is used for all the other measurements in this paper. We measure the spectrum of the excited sublevels by taking absorption images at different excitation frequencies. Figure 1(d) shows an example of such a spectrum in a near electric-field-free environment ($< 1 \text{ V cm}^{-1}$). Our light polarisation allows for transitions with $\Delta m_J = 0, \pm 2$, so that starting from the stretched ground state ($m_J = 1/2$) we address $23D_{3/2}$ with $m_J = -3/2, 1/2, 5/2$. The transitions to $m_J = -5/2, -1/2, 3/2$ are also visible, due to imperfect polarisation and remaining electric fields, albeit suppressed relatively to the others.

3. Results

3.1. Measuring stray electric fields

The future goal of the experiment is to excite atoms to a Rydberg state in traps only 10 $\mu$m from the atom chip surface [22, 28], which requires a good knowledge of the local stray electric fields. We probe these fields by positioning the cloud at different distances to the surface and measuring the excitation spectrum to the $23D_{3/2}$ state. Figure 2 shows these spectra at different distances. Upon approaching the surface, the depletion peaks are shifted to negative frequencies, and a reduction and broadening of the spectrum is visible. Both these effects suggest increasing electric fields and increasing electric field gradients with decreasing distance from the surface.

To better examine the underlying mechanism, we use the fact that we can compensate the electric field $E_z$ in the $z$-direction by applying a homogeneous field $E_{app}$ using the in-vacuum lens covered with indium–tin oxide. The minimum frequency shift of the spectrum occurs when $E_z$ is effectively cancelled by $E_{app}$, so we take spectra for different applied electric fields. Figures 3(a) and (b) show two measurements using the $23D_{3/2}$ state at two different heights. To extract realistic field values from these measurements, we evaluate the atomic transition frequencies for a given magnetic $B_{HP}$ and electric field. The magnetic field is measured independently and is directed predominantly in the $x$-direction (we neglect

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{Depletion spectra for the $23D_{3/2}$ Rydberg state at different distances from the atom chip surface. We plot the normalised depletion $1 - \frac{N_{hole}}{N_{ref}}$ as the colour scheme (colour bar is linear here and throughout the paper), where $R = 0.77$ is the ratio between $N_{hole}$ to $N_{ref}$ without the excitation lasers. The distance to the surface, represented as vertical shift between different curves, is taken from magnetic potential calculations which are calibrated to the experiment. Already at a distance of 169 $\mu$m there is a shift of $\sim 10$ MHz compared to the transition frequency presented in figure 1(d), which was obtained in a nearly field-free environment after using UV-light. In addition, comparing the absorption at 134–98 $\mu$m the spectrum almost completely disappears after a change of only 36 $\mu$m, together with a significant broadening.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3.png}
\caption{Depletion spectrum of the $23D_{3/2}$ Rydberg state taken at distances of (a) 169 $\mu$m and (b) 134 $\mu$m from the surface for different applied fields $E_{app}$ in the $z$-direction, showing a clear dependency of the overall frequency shift of the spectrum on the applied field. The solid lines are theoretical predictions for the peak positions based on the method described in the text, where the stray field parameters have been chosen to best match the observed spectral dependency on $E_{app}$. (c) Extracted stray field values in the $z$-direction, together with a fit based on the electric field of a Gaussian patch of Rb adatoms (see text).}
\end{figure}
the small tilt between $B_{\parallel}$ and the $x$-direction visible in figure 1(c). We assume a linear Zeeman shift for the $m_J$ states and write the Zeeman Hamiltonian for a quantisation axis defined by the total electric field, constituted by $(E_x, E_y, E_z = E_{app})$. The Stark shifts are taken from the diagonalisation of the Stark–Hamiltonian in a sufficiently large set of Rydberg states that are close in energy. Subsequently we diagonalise the combined Zeeman–Stark Hamiltonian to obtain the atomic energies. If we plot these energies as a function of the applied field $E_{app}$, we can well reproduce the peak positions in the experimental spectra for a given set of stray electric fields $(E_x, E_y, E_z)$. For example, the measurement in figure 3(a) yields $(E_x, E_y, E_z) = (5, 8, 22) \text{V cm}^{-1}$. We find a similar value for $E_z$ by an independent measurement for $25S_{1/2}$.

The extracted stray field values $E_z$ are plotted in figure 3(c), which shows a strong increase in electric field from 22 to 45 V cm$^{-1}$ with decreasing distance to the surface. The observed fields are about one order of magnitude larger than what was found in a previous chip experiment with a plain Au-surface [19]. Our measurement shows that the stray fields’ $z$-component points away from the surface. This is consistent with a field induced by Rb adatoms [19, 21]. The electric field above the centre of a Gaussian patch of Rb adatoms can be described in the $z$-direction as [19]

$$E_z = \frac{d_0}{2\omega_0^2} \left[-Z + e^{-2Z^2} \frac{\sqrt{\pi}}{2} \left(1 + Z^2\right) \text{Erfc} \left(\frac{Z}{\sqrt{2}}\right)\right]$$

with the $e^{-1/2}$ patch radius $w$, $d_0$ the peak dipole density, Erfc the complementary error function, and $Z = z/w$. When we fit this expression to our data in figure 3(c), we retrieve a patch radius $w = 70 \text{ nm}$ and a peak dipole density $d_0 = 1.2 \times 10^7$ Debye $\text{nm}^{-2}$. Assuming a dipole moment of 12 Debye per adatom [21], we retrieve an average adatom spacing of 1 nm, which is comparable to the value found in [21], but an order of magnitude smaller than what was found in [19]. The gradient at a height of $134 \text{ nm}$ above the centre of the patch is $\partial E_z/\partial z = 6200 \text{ V cm}^{-2}$.

In order to estimate the $x$-component of the electric field gradient, we further analyse the measurement in figure 2 taken at a distance of $134 \text{ nm}$. In this measurement the 480 nm laser was pulsed for $200 \text{ ms}$ (see section 3.3) and the 780 nm laser for $100 \mu\text{s}$. Instead of defining a region-of-interest (ROI) that includes the entire excitation beam area (the ‘hole’ square in figure 1(c)), we divide this ROI into smaller areas in the $x$-direction, each $10 \mu\text{m}$ wide, and plot the spectrum for the different sub-areas (figure 4). A strong change in transition frequency is visible over only $100 \mu\text{m}$. In order to extract the corresponding field gradient, we use the theoretical description of the atomic energy levels assuming a linear field gradient in the $x$-direction. From that we can extract a gradient value of $\partial E_x/\partial x = 3500 \text{ V cm}^{-2}$, comparable to the value found for the $z$ field gradient. We do not observe a gradient in the $y$-direction, possibly because we are naturally limited by the smaller cloud size in that direction.

Our atom chip features a $200 \text{ nm}$ thick micro-structured FePt layer beneath the Au coating. The FePt layer is magnetised, such that, together with a homogeneous external magnetic field, it creates both a square and hexagonal lattice of magnetic micro-traps close to the surface. Details of the chip design and the trapping procedure are given in reference [22]. In order to obtain data close to the surface ($\sim 10 \mu\text{m}$), where the magnetic trapping potential is influenced by the magnetic micro-traps, we change our excitation scheme to obtain a higher Rabi frequency. Contrary to the rest of the measurements in this paper, we tune our lasers on resonance ($\Delta = 0 \text{ MHz}$) with the intermediate $5P_{3/2}$, $F = 3$ level, leading to a two-photon on-resonance transition with much higher Rabi frequency to the $25S_{1/2}$ state. The imaging laser is used here for the excitation to the intermediate level at $780 \text{ nm}$ with a pulse time of $100 \mu\text{s}$. The $480 \text{ nm}$ laser is on for $200 \text{ ms}$. Figure 5 shows optical density images while changing the frequency of the $480 \text{ nm}$ laser or the applied electric field $E_{app}$. As in figure 1(c) atoms excited to a Rydberg state are lost and appear as a hole in the cloud. The size of this hole is much smaller than the laser beam diameters. The hole thus marks where the excitation is resonant. We use two different trap configurations: a macro-wire trap where the lower part of the atomic cloud is already influenced by the micro-trap potential (figures 5(a)–(f)), and atoms confined in the micro-traps (figures 5(g)–(i)). In those images, a change in excitation frequency results in a spatial translation of the depletion area in the $y$-direction. Similarly, a change in applied field $E_{app}$ leads to a spatial shift in the same direction. This points at strong electric field gradients in the $y$-direction. Remarkably, the transition frequency changes by $100 \text{ MHz}$ over a distance of a few tens of micrometers, corresponding to a gradient of about $\partial E_y/\partial y = 1700 \text{ V cm}^{-2}$. Varying the applied voltage on the lens, the depletion area traverses only half of the cloud, which suggests that we only cancel the $E_y$ with strong fields $E_x, E_y$ still present. For our measurements at that small distance, the spatial depletion changes from shot...
to shot, and the involved frequencies from day to day, making a quantitative analysis unreliable.

3.2. Reduction of the stray electric fields

The presence of strong stray electric fields and gradients near the surface poses an obstacle for exciting Rydberg atoms at the chip. Reducing these fields is crucial to our goal, and we investigate several methods for removing the source of these fields.

One method, as suggested in [21], is to excite all the atoms in the MOT to a Rydberg state, which will lead to subsequent ionisation of a large fraction of the excited atoms. The resulting charges can settle on the surface and compensate for the electric stray field. Contrary to the findings in [21], where a low number of electrons significantly reduces the positive adatom field, we do not see any compensation effect. Looking at the differences between the experiments, a possible explanation might be that we do not have a bulk mono-crystalline layer of SiO₂, and that the exposure of the surface to the excitation lasers might remove the free charges.

In a second attempted method we deliberately deposit large atomic clouds on the surface, as it was found in [6] that a more homogeneous layer of adsorbed atoms can decrease the stray fields. In our case, however, this procedure increases the stray fields.

As a third method, we illuminate the atom chip with UV light at 365 nm as we expect UV light to influence the surface adatoms via the light-induced atomic desorption (LIAD) effect [29, 30]. The UV light is provided by an array of nine 1W LEDs, which is brought in close proximity to the vacuum quartz cell walls, releasing a lot of additional Rb atoms before the excitation pulse, due to LIAD from the vacuum quartz cell walls, releasing a lot of additional Rb atoms. To quantify the effect on the electric stray fields, we perform the same measurements as in figure 3. The results are shown in figures 6(a) and (b), and show a spectrum with small electric stray field at 169 μm, as indicated by the equally spaced depletion peaks. As shown in figure 1(d), the level spacing is consistent with the Zeeman splittings. If we decrease the distance, we find an increasing electric field. In general, even though the data is less well reproduced by our theory after using the LEDs, the $E_z$ value can still be estimated well (compared to $E_x$, $E_y$) from the spectrum. The field in the $z$-direction and a fit to a Gaussian patch model (equation (1)) are plotted in figure 6(c), showing a strong reduction compared to figure 3(c), although the fields are still larger than those of [19]. We can also conclude from the poor fit that the Gaussian patch model no longer represents the underlying adatom distribution.
to the surface. The electric stray field is measured by applying an external field, \( E_{\text{app}} \), and measuring the excitation spectrum, as in figure \( \text{6} \). The decay time of the number of atoms is equal to the increase time of the stray fields, \( \sim 1 \text{ d} \). The data marked with an open blue circle is not included in the number of atoms fit because it belongs to a dataset with a colder, hence smaller, cloud. The corresponding measurement of the stray fields is included in the stray fields fit because the field measurement is independent of the number of atoms.

3.3. Temporal behaviour of the stray fields

In order to better understand the influence of the UV light on the stray fields, we look at the temporal dynamics of this effect. Within one day of illuminating the cell and the atom chip with UV light continuously, leading to a strong initial reduction of stray fields, the number of atoms starts to decrease and simultaneously the stray fields increase. The reduction of the number of atoms indicates that the UV illumination cleans the inside of the vacuum quartz cell from Rb atoms accumulated there. However, it is unclear why this leads to an increase of the stray fields. Figure \( \text{7} \) shows the reduction of the number of atoms after the evaporation stage and the corresponding increase of the stray electric field during the first week of operating the LEDs, at a height of \( 134 \mu \text{m} \). Both exponentials have the same time constant of \( \sim 1 \text{ d} \), suggesting a negative correlation between number of atoms and stray fields under our operating conditions. After a week of operating the LEDs, the decrease in number of atoms does not leave sufficient atoms in the magnetic trap for taking spectra. When we increase the background pressure again by switching off the LEDs and operating the system normally for several hours, we can retrieve the original number of atoms. If we then switch on the LEDs we see the same increase in number of atoms and reduction of stray fields. At longer time scale the number of atoms drops and the stray fields increase, as in figure \( \text{7} \).

In order to show that the reduction of stray electric fields due to UV light occurs within one experimental cycle (\( \sim 20 \text{ s} \)) after switching on the LEDs, we use the following procedure: (i) build up Rb pressure by normally operating the system without LEDs, usually for several hours, (ii) tune the excitation lasers to the low-field transition frequency as measured in figure \( \text{6} \), yielding no depletion at all, and (iii) turn on the UV light. In this procedure, the depletion becomes visible only if the stray fields decrease. Figure \( \text{8} \) is the result of such a procedure, where the LEDs are turned on in the beginning of experimental cycle \( 0 \). An immediate reduction of the stray fields in one experimental cycle is visible. After a few more experimental cycles we had to stop the measurement in order to rebuild the Rb pressure.

The temporal dynamics shown in figures \( \text{7} \) and \( \text{8} \) suggest that the dominant effect of the UV light is not the removal of adatoms from the surface itself, as the stray fields increase again while the UV light is present. We speculate that we influence the adatom distribution by increasing the Rb background pressure thereby creating a more uniform or larger layer, resulting in a decrease of stray fields.

We also examine the influence of the 480 nm excitation laser on the stray fields. The laser hits the atom chip surface at normal incidence with a \( \sim 40 \mu \text{m} \) 1/e² radius. We estimate that permanent exposure with that laser increases the local surface temperature by \( \sim 50 \text{ K} \) (this number is based on our laser intensity, surface reflectivity, and the thermal conductivities of the Au film, the SU8 layer, and the Si substrate). This temperature increase can cause desorption of adatoms which reduces stray fields as shown in \( [21] \). In addition, we expect an effect due to LIAD, as the local light intensity is orders of magnitude higher than for the LED array. If the surface is exposed to the laser for several seconds after the excitation pulse and the imaging sequence, there is no change of the stray fields in the next shot (\( \sim 15 \text{ s} \) delay). On the other hand, the effect due to LIAD is minor compared to the effect due to the UV LEDs.
hand, if the surface is illuminated before the excitation pulse, we measure a reduction of the electric stray field, which increases with the exposure time, as shown in figure 9.

This reduction is also present in figures 4 and 5 compared to figure 3, as those measurements were taken with 200 ms long 480 nm pulses. The minimum stray field in the x-direction in figure 4 roughly corresponds to the beam centre. Furthermore, in figure 5 the depletion is only visible in the beam centre. The electric stray fields corresponding to the detuning of $-116$, $-76$ and $-36$ MHz in figures 5(a)-(c) are $25$, $20.25$ and $14$ V cm$^{-1}$, which is significantly lower than the fields in figure 3(c).

4. Discussion

In [17] electric surface fields were measured $\approx 10 \mu$m from different surfaces by a change in oscillation frequency of a Bose–Einstein condensate. The fields were attributed to Rb adatoms on the surface. When BK7 glass is used as surface material, the stray fields are significantly lower compared to Si or Ti surfaces, less than $1$ V cm$^{-1}$. Similarly low electric fields were found in [31] where fused silica was used as surface material. A low field was also found in micro-metre size glass vapor cells [32]. In contrast to this, measurements with Yttrium [18] and Au [19, 20] show larger stray fields which increase with the amount of deposited Rb on the surface. In comparison, this seems to suggest that a dielectric surface is superior to a metal surface in terms of stray fields. However, for our SiO$_2$ coating we find stray fields which are roughly one order of magnitude larger than for our earlier atom chip with an Au surface [19]. Our finding is consistent with the measured stray fields found in [21] for SiO$_2$. Furthermore, as the sign of the electric dipole moment can be used to distinguish between chemisorbed and physisorbed adatoms [5], our measurements supports chemisorbed adatoms compared to a weaker binding due to van der Waals forces as assumed in [17]. This again is consistent with [21].

In contrast to the measurements in [21], we do not see a decrease of stray fields when producing free charges by Rydberg excitation in the MOT. This might be related to two differences in the experiments: we do not have a monocrystalline bulk of SiO$_2$, and our 480 nm laser impinges directly on the surface. Both effects might interfere with the compensation effect due to a small number of surface electrons observed in [21].

We do see an immediate improvement after exposing the surface and vacuum cell to UV light. The temporal dynamics of that effect suggest that rather than directly removing adatoms from the surface, the UV light creates a more uniform layer of Rb atoms by transferring atoms from the vacuum windows to the chip surface. Such a uniform layer was used in [6] to lower the stray fields. In our case the increase in Rb background pressure might increase the adsorbate coverage on the surface [5]. This is also consistent with the observation that the stray field increases again with decreasing background pressure and number of atoms in the magnetic trap.

Finally, the 480 nm laser clearly lowers the local electric field around its contact area with the surface. This suggests that, in contrast to the UV light, we desorb adatoms, either by LIAD or by a local increase in temperature that was found to lower the stray fields [5, 18, 21]. It is important to note that the lowering of the stray fields is only visible if the surface is exposed to the 480 nm laser immediately before the excitation pulse. Most likely Rb atoms produced in the next experimental cycle replenish the desorbed adatoms.

5. Conclusions

We have found that a coating of SiO$_2$ on our Au surface significantly increases the electric stray field compared to a plain Au surface. We can reduce the stray field by using UV-light, but at the expense of the number of atoms in the magnetic trap. It may thus be possible to strike a balance between these two effects and find an optimum working point. Whereas [21] shows that a significant decrease of the stray field is possible for SiO$_2$ as a bulk material, we could not achieve this improvement, and it is unclear if this is possible for micro-traps in close proximity to the surface ($\approx 10 \mu$m) and in the presence of a high-power 480 nm excitation laser. Our results also suggest that heating the surface by a few tens of K is a viable option to significantly reduce the stray fields.

The fine interplay between the Rb atoms being adsorbed on the chip surface, the Rb atoms on the vacuum glass cell, and the influence of the UV-light and the temperature-increase causes both fast (single-cycle) and slow (~day) changes that need to be carefully considered in future experiment.
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