Transient Density-Induced Dipolar Interactions in a Thin Vapor Cell

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We exploit the effect of light-induced atomic desorption to produce high atomic densities \( n \gg k^3 \) in a rubidium vapor cell. An intense off-resonant laser is pulsed for roughly one nanosecond on a micrometer-sized sapphire-coated cell, which results in the desorption of atomic clouds from both internal surfaces. We probe the transient atomic density evolution by time-resolved absorption spectroscopy. With a temporal resolution of \( \approx 1 \) ns, we measure the broadening and line shift of the atomic resonances. Both broadening and line shift are attributed to dipole-dipole interactions. This fast switching of the atomic density and dipolar interactions could be the basis for future quantum devices based on the excitation blockade.

The effect of dipole-dipole interactions in optical media becomes important when the density \( n \) is significantly larger than the wave number cubed \( k^3 \) of the interaction light field. Entering this regime leads to interesting nonlinear effects such as an excitation blockade [1], nonclassical photon scattering [2], self-broadening (collisional broadening) [3], and the collective Lamb shift [4, 5]. Dipole-dipole interactions are observable in steady-state experiments performed in thin alkali vapor cells [6–8], where the cells are heated to temperatures above 300 °C. Dipolar broadening effects were previously observed to be independent of the system geometry, while the line shift depends on the dimensionality of the system, as investigated in a 2D model [8, 9]. It is however not straightforward to prepare high densities with alkali vapors [10]. One technique to increase the atomic density is light-induced atomic desorption (LIAD) [11–16] or light-activated dispensers [17]. LIAD is commonly used for loading magneto-optical traps [18–20] and has been studied in confined geometries like photonic-band gap fibers [21, 22] or porous samples [23]. However, the application of pulsed LIAD for fast switching of dipole-dipole interactions is so far unexplored.

In our pulsed LIAD setup, we can switch the atomic density in the nanosecond domain, which allows one to study the dipole-dipole interaction on a timescale faster than the natural atomic lifetime. This fast density switching has been already used in our group to realize an on-demand room-temperature single-photon source based on the Rydberg blockade [24]. In this work, we study the dipolar interaction for the two transitions \( D_1 : 5S_{1/2} \rightarrow 5P_{1/2} \) and \( D_2 : 5S_{1/2} \rightarrow 5P_{3/2} \) of rubidium with different transition dipole moments.

We first describe our LIAD measurement results in a thicker part of the cell [cell thickness \( L = 6.24(7) \) μm] at a low density \( (nk^{-3} \approx 1) \). This measurement is used as the basis to set up a model for the velocity and density distribution of the desorbed atoms. Then, we focus on a thinner part of the cell \( [L = 0.78(2) \) μm], where we can study transient density-dependent dipolar interactions at a high density (up to \( nk^{-3} \approx 100 \)). To this end, we compare two transitions \( (D_1 \) and \( D_2 \) transition of rubidium) with different transition dipole moments to investigate their influences on the dipole-dipole interaction in a quasi-2D geometry \([L \approx \lambda_{\text{probe}}] \).

We use a self-made, wedge-shaped, micrometer-sized cell with an attached reservoir tube filled with rubidium (72% \(^{85}\)Rb, 28% \(^{87}\)Rb), shown in Fig. 1(b). By heating the cell independently from the reservoir, we can produce a certain rubidium coverage on the cell walls and a vapor pressure in the cell with a comparably small background density \( n \approx 10^{14} \) cm\(^{-3} \) (reservoir temperature \( T_{\text{res}} \approx 180 \) °C). In our setup, the pulsed LIAD laser and the probe laser are aligned collinearly in front of the cell [Fig. 1(a)]. The pulsed LIAD laser at 532 nm has a pulse length of 1.1(1) ns (FWHM) and a repetition rate of 50 kHz. This off-resonant pulse leads to the desorption of rubidium atoms bound to the sapphire-coated glass surface. The amount of desorbed atoms depends on the peak intensity \( I \) of the LIAD pulse. We probe the desorbed atoms at 795 nm \( (D_1 \) transition of Rb) or 780 nm \( (D_2 \) transition of Rb). Both probe lasers have an intensity well below the resonant \( D_2 \) saturation intensity \( I_{\text{sat,D}_2} < 0.01 I_{\text{sat,D}_1} \). The measured Gaussian beam waist radius \( (1/e^2) \) of the probe laser is \( w_{\text{probe}} = 2.0(2) \) μm, while the LIAD laser has a waist radius of \( w_{\text{LIAD}} = 13.7(1) \) μm. We measure the transmitted photons with a single-photon counting module, which is read out by a time tagger module. Our wedge-shaped cell has a point of contact of the cell walls, which can be seen...
The desorbed atoms are emitted with a certain velocity distribution into the cell volume from both sides of the cell, where they are probed with a red laser (probe laser). A map of the time- and detuning-resolved ∆OD depth caused by the background vapor pressure is sub-
tracted. A map of the time- and detuning-resolved ∆OD is shown in Fig. 2(a). At $t = 0$ ns the LIAD pulse hits the cell and increases the optical depth. The time resolution of the measurements is limited by the time jitter of the LIAD pulse (500 ps) and the single-photon counting module (350 ps).

First, we focus on a thicker part of the cell [$L = 6.24(7) \mu$m], where we measure a time- and detuning-
resolved ∆OD map of the $^{85}$Rb $F_g = 2$ transition of the $D_2$ line [Fig. 2(a)]. The transition is defined by the total angular quantum number $F_g$ of the ground state $g$, while the total hyperfine splitting $\delta_{\text{hfs}}$ of the excited state cannot be resolved due to transient and Doppler broadening. The atoms moving in the laser propagation direction, originating from the entry facet of the cell, are probed at blue detunings $\delta > 0$. A second group of atoms, originating from the exit facet, is visible and probed at red detunings $\delta < 0$. The signal is higher for the atoms moving in the laser propagation direction. This asymmetry is not anticipated, but might originate from differences in the surface properties as it was observed in other experiments, i.e., depending on the coating [21]. We checked this hypothesis by rotating the cell by 180°, which led to a roughly inverted asymmetry. The darker region around zero detuning shows that fewer atoms with low $z$ velocity are desorbed. In total, we measure two atom clouds moving toward the opposite cell walls. The high ∆OD value in the first nanoseconds is caused by a high atomic density and decreases over time. The ∆OD signal equilibrates to zero before the next LIAD pulse arrives. The dashed green lines indicate the time-of-flight curves, after which the atoms with a certain detuning hit the other cell wall according to $\text{TOF}(\delta) = Lk/|\delta|$, with $\delta = k \cdot v \pm \delta_{\text{hfs}}/2 = k v_z \pm \delta_{\text{hfs}}/2$, respecting the hyperfine splitting of the excited state. There, $k$ is the wave number of the probe beam, which is parallel to the $z$ axis, $k = |k|$ is the wave number of the probe beam, $v$ is the velocity of the atom, and $v_z$ is the $z$ component of the velocity. We observe distinct signal wings beyond the respective time-of-flight curves indicating potential re-emission of

![Figure 1](image1.png)

**FIG. 1.** (a) Illustration of the LIAD process. A green laser pulse (LIAD pulse) desorbs rubidium (Rb) atoms, which are adsorbed on the inner, sapphire-coated surface of the glass cell. The adsorbed atoms are emitted with a certain velocity distribution into the cell volume from both sides of the cell, where they are probed with a red laser (probe laser). (b) Photo of the wedge-shaped micrometer-sized cell with interference fringes (Newton’s rings). The cell thickness in the probe region is determined interferometrically and ranges from 0.78(2) to 6.24(7) µm from left to right.

![Figure 2](image2.png)

**FIG. 2.** (a) Measured ∆OD map of the $^{85}$Rb $F_g = 2$ transition of the $D_2$ line. After the atoms, depending on their detuning ($z$ velocity), hit the other cell wall the signal decreases. The TOF(δ) is shown with two dashed green lines. The intensity $I$ is the peak intensity of the LIAD pulse. The cell thickness is $L = 6.24(7) \mu$m and the reservoir temperature is $T_{\text{res}} \approx 140$ °C. (b) Simulated ∆OD map of desorbed atoms. The simulation parameters are adapted to the measurement. The TOF(δ) curves (dashed green lines) are plotted, too. The indicated $n_p$ is the peak density at 2 ns.
atoms after arriving at the opposite cell wall. Using this measurement as a reference, we develop a kinematic model and run a Monte Carlo simulation of atoms flying through a cell and interacting with the probe laser (see Supplemental Material [25]). The idea is to model the velocity distribution of desorbed atoms and to estimate the local density during the simulation. In the model, the local and temporal desorption-rate scales linearly with the intensity of the LIAD pulse. For the velocity distribution we assume \( f(v, \varphi, \theta) = \frac{4}{\sqrt{\pi}a^3}v^2 \exp(-v^2/a^2) \cos(\theta) \) with the parameter \( a \) and the speed \( v = |v| \). The azimuthal angle \( \varphi \) is uniformly distributed, while the polar angle \( \theta \) is distributed according to the \( \cos(\theta) \)-Knudsen law [31]. This simple distribution leads to a good qualitative agreement between measurement [Fig. 2(a)] and simulation [Fig. 2(b)]. We also assume that the atoms are desorbed only during the LIAD pulse and that there is no thermal desorption after the pulse, which is in good agreement with our measurement. Since no other mechanism (i.e., through natural- or transit-broadening, which are also included in the model) reproduces the signal wings beyond the time-of-flight curves (dashed green lines in Fig. 2), they might occur because of re-emissions from the surfaces after bombardment with the initial atom clouds. To get better agreement, an instant re-emission probability of 84\% is included in the kinematic model. The remaining discrepancies between measurement and simulation can originate from an inadequate velocity distribution model, intricate re-emission properties, additional decay mechanisms, the neglected Gaussian intensity distribution of the probe beam, and the use of the steady-state cross section of the atoms at all the times. Nevertheless, with the overall acceptable agreement between measurement and simulation we obtain a time- and \( z \)-dependent simulated local density, which shows that the desorbed atoms are initially in two flat, "pancake-like" clouds with an initial thickness well below the wavelength of the probe laser, rendering this into a 2D geometry (see Supplemental Material [25]).

To investigate high-density regimes, we use a thinner cell as the background optical depth and the detection limit of the single-photon counting module are limiting the measurement in the thicker part of the cell. We perform measurements at a cell thickness of \( L = 0.78(2) \) \( \mu \)m at low and high atomic densities using the \( D_2 \) transition as shown in Figs. 3(a) and (b), respectively. Our measurements are in a regime where the total number of desorbed atoms per pulse monotonically increases with the peak intensity of the LIAD pulse (see Supplemental Material [25]). The low-density measurement corresponds to a peak intensity of \( I = 2.6(3) \) MW cm\(^{-2}\), while the high-density case corresponds to \( I = 211(21) \) MW cm\(^{-2}\). There is a broadening and line shift of the \( D_2 \) hyperfine transitions present in Fig. 3(b), which we attribute to density-dependent dipole-dipole interactions. The four peaks correspond to the ground state hyperfine splitting of the two isotopes of rubidium, contributing to the signal, while the hyperfine splitting of the excited state can not be resolved. The density-dependent self-broadening [3, 6] in the steady-state regime was predicted to be

\[
\Gamma_{\text{self}} = \beta_i n = \frac{2}{3h\epsilon_0} \sqrt{\frac{g_g}{g_e}} d_J^2 n ,
\]

where \( \beta_i \) is the self-broadening coefficient, \( i \) enumerates the \( D_1 \) or \( D_2 \) transition, \( h \) is the reduced Planck constant, \( \epsilon_0 \) is the vacuum permittivity, \( g_g \) and \( g_e \) are the multipoleities (depending on the quantum number \( J \)) of the ground and excited state respectively, \( d_J \) is the total reduced dipole matrix element, and \( n \) is the atomic density. In the high-density regime in Fig. 3(b) we observe a self-broadening of \( \Gamma_{\text{self}} \approx 590 \Gamma_0 \) at \( t = 2 \) ns, where \( \Gamma_0 \approx 2\pi \times 6.07 \) MHz [32] is the natural decay rate of the \( D_2 \) transition.

Similarly, we compare the line shift, observed in our measurements, to the steady-state dipole-dipole shift [5, 7], which was predicted to be

\[
\Delta_{dd} = -|\Delta_{LL}| + \frac{3}{4} |\Delta_{LL}| \left( 1 - \frac{\sin 2kL}{2kL} \right) ,
\]

with \( \Delta_{LL} \) being the Lorentz-Lorenz shift and \( L \) being the cloud thickness. This thickness dependency is a cavity-induced correction, also known as the collective Lamb
shift. The Lorentz-Lorenz shift [5, 7], in turn, is density-dependent and can be written as

$$\Delta_{LL} = -\frac{1}{3\hbar c_0} d_j^2 n.$$  \hspace{1cm} (3)

As our cell thickness is $L \approx \lambda$, the second term of the dipole-dipole shift has a significant effect on the line shift and reduces the dipole-dipole effect to $\Delta_{dd} \approx -\frac{1}{3} |\Delta_{LL}|$. In the high-density measurement in Fig. 3(b) this corresponds to a value of $\Delta_{dd} \approx -80 \Gamma_0$ (redshift) at $t = 2$ ns. Additionally, we can observe that the transient density-dependent effects occur on a timescale of a few nanoseconds, which is faster than the natural lifetime of the $D_2$ transition (26.2 ns) [32].

To further investigate the dipole-dipole origin of the observed interaction, we compare the transient evolution of the self-broadening and line shift at the $D_1$ and $D_2$ transition of rubidium. We fit both measured data with a steady-state electric susceptibility model at each time step, using the software ElecSus [33]. The fits to the individual time-resolved spectra show a < 6% overall normalized root-mean-square deviation and result in the self-broadening and line shift shown in Figs. 4(a) and (b), respectively. Note that in the first 2 ns we cannot properly fit the data to this model, so we exclude these data points. The error bars represent the 1σ standard fit error (see Supplemental Material [25]). If we assume that the self-broadening and the line shift, according to the aforementioned steady-state equations, linearly depend on the density, we can calculate a peak density on the order of 10^{16} cm^{-3} using Eqs. (1) and (2).

There is an apparent difference of the self-broadening and line shift between the two transitions of rubidium, which can be attributed to different transition dipole matrix elements $d_{jj}$. While it is not possible to conclusively deduce any precise value for $d_{jj}$ from our data, we calculate ratios between the $D_1$ and $D_2$ broadening and shift for otherwise identical measurements, which are shown in Figs. 4(a) and (b) on the right vertical axis. These values approach the ratios $\sqrt{\frac{1}{J^2} d_{J,D_2}^2/d_{J,D_1}^2}$ and $\frac{d_{J,D_2}^2}{d_{J,D_1}^2}$ emerging from Eqs. (1) and (2), respectively, for large $t$ as indicated by the black triangles. Deviations during the first $\approx 10$ ns in the case of the self-broadening likely originate from limited accuracy of the fits with signal wings not captured with the scanned detuning range, asymmetries in the spectral profiles similar to what was reported in Ref. [8], or asymmetries from both hyperfine splitting and velocity distribution (see Fig. 5, Supplemental Material [25]). In contrast, the measured line shift is always much smaller than the scan range while almost vanishing for $t > 8$ ns such that the error bars are larger than the values themselves. Such systematic uncertainties are not properly captured by the standard errors as derived from the employed fitting algorithms.

In conclusion, we implemented a pulsed LIAD method to switch atom densities from 10^{14} cm^{-3} to more than 10^{16} cm^{-3} on a nanosecond timescale in a micrometer-sized cell. At high densities with $nk^{-3} \approx 100$ we are able to study the dipole-dipole induced self-broadening and Lorentz-Lorenz shift. Our measurements show that the interaction builds up faster than 2 ns, a timescale much shorter than the natural lifetime. The scaling between the $D_1$ and $D_2$ transition in the measurement supports the assumption that we observed dipolar effects in good agreement with the established theory. Overall, we do not see significant transient internal dynamics other than the one induced by the density change itself, since the motional dephasing is the fastest timescale equilibrating the shift and broadening of the many-body dynamics with dipolar interactions within $\approx 1$ ns. With a better temporal resolution (e.g., with superconducting single-photon detectors) and shorter desorption pulses, it will be possible to study the behavior of the transient dipole-dipole interaction in the first 2 ns, a regime which was not accessible in this work. The switching of the atomic medium

![FIG. 4. (a) Time-dependent self-broadening $\Gamma_{\text{self}}$ of the Rb spectra for the $D_1$ (yellow data) and $D_2$ (purple data) transition. The experiments are performed under identical conditions, except the probe laser wavelength. The ratio (green data) of the self-broadening of the two transitions approaches the theoretical steady-state ratio for large $t$ (black triangle). Vertical error bars for $t < 8$ ns are likely underestimated due to a systematic effect, and horizontal precision is bandwidth limited by jitter. (b) Time-dependent line shift $\Delta_{sm}$ of the $D_1$ and $D_2$ transition. The ratio of the line shift, which is close to the theoretical value, has an increasing error for increasing time, and therefore the last three data points were omitted. The peak intensity of the LIAD pulse is $I = 137(3)\text{ MW cm}^{-2}$. The cell thickness is $L = 0.78(2)\mu m$ and the reservoir temperature is $T_{res} \approx 180^\circ C$.](image-url)
by LIAD can be used with integrated photonic structures [34, 35], e.g., to realize large optical nonlinearities at a GHz bandwidth for switchable beam splitters, routers, and nonlinear quantum optics based on the excitation blockade.

The supporting data for this article are openly available from [36]. Additional data (e.g., raw data of the time tagger) are available on reasonable request.

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SUPPLEMENTAL MATERIAL FOR
“TRANSIENT DENSITY-INDUCED DIPOLAR INTERACTIONS IN A THIN VAPOR CELL”

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I. OVERVIEW AND DEFINITIONS

This Supplemental Material roughly follows the structure of the manuscript, providing additional information alongside. In section II the experimental prerequisites to perform our LIAD measurements are detailed and the principle data analysis procedure is introduced. Section III elaborates our findings about the behavior of the desorption process when the LIAD pulse intensity is increased. This forms a necessary foundation for the development of our kinematic model discussed in section IV, as the latter makes certain assumptions about the number of atoms which get desorbed during each pulse. Section V frames the discussion of Fig. 4 in the manuscript by showing how the individual data points were processed and compared. Section VI concludes this work by highlighting another approach pursued to deepen the understanding of the dipolar origin of the observed effects. This Supplemental Material and the manuscript share consistent definitions of all introduced symbols. All frequencies are given as angular frequencies, such that detunings \( \Delta \) and decay-induced linewidths \( \Gamma \) all appear on the same scale without additional conversion factors. We define the laser detuning \( \delta \) as the actively adjusted laser frequency \( \omega_l \) compared to a resonant atomic transition frequency \( \omega_a \) like \( \delta = \omega_l - \omega_a \). An additional detuning occurs due to the velocity \( v \) of the atoms as we deal with a thermal gas. Moreover, there are additional sources of line shifts such as dipole-dipole effects \( \Delta_{dd} \) which are included in the perceived detuning \( \Delta_{atom} \) of an atom given by

\[
\Delta_{atom} = \omega_a - \omega_l + k \cdot v + \Delta_{dd} \quad (S4)
\]

\[
= -\delta + k \cdot v + \Delta_{dd} \quad (S5)
\]

The time-of-flight TOF(\(\delta\)) curves in Fig. 2 in the manuscript and Fig. S3, which are given in the maps as an guide to the eye, can be understood as the time-of-flight solution of Eq. (S5) for an atom with zero perceived detuning \( \Delta_{atom} = 0 \) and no additional shifts \( \Delta_{dd} = 0 \) (i.e. resonant interaction under Doppler effect with otherwise negligible linewidth) when traveling across the cell.

II. EXPERIMENTAL SETUP

In order to observe a time- and detuning-resolved LIAD process and the consequent velocity and density map, each detected photon must be assigned to a time \( t \) after the LIAD pulse and a detuning \( \delta \). We use a single-photon counting module (SPCM-AQRH-10-FC from Excelitas), which has a specified single photon timing resolution of 350 ps. Its electrical triggers are temporally digitized by a time tagger module (Time Tagger 20 from Swabian Instruments) with a specified RMS (root-mean-square) jitter of 34 ps. This produces an absolute timestamp \( t_{SPCM,k} \) for each observed photon \( (k \in \mathbb{N}) \).

The LIAD laser pulses are electrically triggered from a pulse generator (Bergmann BME_SG08p) at 50 kHz repetition rate with a specified delay resolution of 25 ps and an output to output RMS jitter of less than 50 ps. These electrical pulses are fed into a Q-switched 532 nm laser (BrightSolutions WedgeHF 532) with an experimentally determined pulse-to-pulse jitter of less than 500 ps. We use an additional synchronized output from the pulse generator as temporal reference for the laser pulses generating additional timestamps \( t_{LIAD,\ell}, (\ell \in \mathbb{N}) \).

At the same time, a Littrow-configuration, external-cavity diode laser (Toptica DL Pro) is continuously
frequency-scanned over the ranges shown in the figures of the manuscript at a rate of 11 Hz. This ensures that the effective detuning $\delta$ is quasi constant during each individual LIAD pulse and the transient regime, which is on the order of less than a microsecond. It additionally provides a large number of LIAD pulses for each detuning in every dataset spanning over an integration time in the order of one day. The scan ramp triggers are fed into the time tagger module and generate a series of timestamps $t_{\text{scan},m}, (m \in \mathbb{N})$. This overall trigger sequence for a single photon detection is schematically depicted in Fig. S1. During analysis, time differences

\begin{align}
\Delta t_{\text{LIAD},k} &= t_{\text{SPCM},k} - t_{\text{LIAD},t} \quad \text{and} \quad (S6) \\
\Delta t_{\text{scan},k} &= t_{\text{SPCM},k} - t_{\text{scan},m} \quad (S7)
\end{align}

are calculated, such that each SPCM timestamp gets assigned to the most recent LIAD and scan trigger. The value $\Delta t_{\text{LIAD},k}$ reflects the timing with respect to the desorption pulse. The different electrical and optical propagation times are corrected by observing LIAD pulses directly on the same SPCM and subtracting their average peak position to obtain time differences on a time axis $t$, which is plotted in all figures in this work. Similarly, $\Delta t_{\text{scan},k}$ relates to the temporary probe laser frequency $\omega_{t,k}$ by a certain function $\omega_{t}(\Delta t_{\text{scan}}, t_{\text{SPCM}})$. The direct dependency on the absolute time $t_{\text{SPCM}}$ indicates that laser frequency drifts must be corrected. This is achieved by calibrating to an ultra low expansion (ULE) cavity captured continuously on a digital oscilloscope. The ULE cavity has a free spectral range of $2\pi \times 1.5$ GHz and an overall negligible drift. The absolute frequency with respect to the atomic transitions can be determined by additionally observing a saturated rubidium spectrum on the same oscilloscope. All values $\delta$ are given with respect to the center-of-mass of the shown transition(s). Note that the function $\omega_{t}(\Delta t_{\text{scan}}, t_{\text{SPCM}})$ is calculated stepwise to reflect the different behavior during the rising and falling scan ramp. We estimate an overall statistical uncertainty for $\delta$ of less than $2\pi \times 10$ MHz, mainly limited by fluctuations of the scan rate during each scan ramp not captured by the $2\pi \times 1.5$ GHz ULE cavity.

The mapped plots with respect to $t$ and $\delta$ are calculated by binning relevant pairs $(t, \delta)_k$ into a 2D histogram. A bin width of $2\pi \times 10$ MHz to $2\pi \times 50$ MHz for $\delta$ and 500 ps for $t$ is used throughout this work. The bin width of $t$ is thus chosen on the same order of magnitude as the time jitter limiting the overall measurement. The binning of the probe laser detuning can be chosen more arbitrarily to suppress the noise. For the displayed data we use a bin-width larger than the free-running linewidth of the probe laser ($\nu \approx 2\pi \times 800$ kHz).

From the various measurements shown in the manuscript and the experience with our setup, we can state, that the observations due to the LIAD effect are repeatable after months and years. Yet we have to mention, that during measurements with high LIAD pulse intensity, the cell is locally modified. This modification manifests itself as a decreased transmission or visibly brown spot, which, depending on the total exposure, can be either temporary and healed by uniform heating or stay permanently. We attribute this behavior to an alteration of the sapphire coating after bombardment with rubidium atoms. We could not observe such a change in transmission in a similar cell filled with air under otherwise identical conditions. We also note that a similar discoloration and loss of transmission is a well-known effect for cells filled with alkali vapor at elevated temperatures [26]. It could be possible that the LIAD effect and velocity distribution locally produce a similar phenomenon even at lower temperatures. To minimize the impact of such effects on our measurements, we monitor the probe transmission and regularly move the cell to a spot with the same cell thickness to continue the measurement once a noticeable transmission loss is observed. Thereby, we gain reproducible results over multiple measurement runs.

### III. Atomic Density for Increasing LIAD Intensity

In this work we present the change of the optical depth $\Delta \text{OD}$ as an indicator for the temporally-controlled, additional atomic density (or equivalently the number of desorbed atoms) compared to a vapor cell homogeneously heated to a certain background temperature. It is plausible to assume that for LIAD pulses of fixed duration, the number of desorbed atoms increases with increasing LIAD pulse peak intensity $I$, since the total energy introduced into the system increases. While we could not deduce a simple relationship between the parameters for a velocity distribution model (see section IV) and the LIAD pulse parameters, we still observe well-defined relationships between $I$ and key features of the corresponding $\Delta \text{OD}$ maps.

One such feature is the peak optical depth value reached on resonance, $\Delta \text{OD}_{\text{peak}}$, as an indicator for the total number of atoms desorbed during the process. Fig. S2(a) shows that these values monotonically increase for both the $D_1$ and $D_2$ transition with increasing $I$. As the attainable atomic density is independent of the probe wavelength and therefore roughly identical in both cases, the ratio between the individual $\Delta \text{OD}_{\text{peak}}$ values for both transitions should be proportional to their resonant scattering cross section $\sigma_0$, which in turn can be related to the transition dipole moment $d$ as $\sigma_0 \propto |d|^2$ [27]. This assumes steady-state behavior and is therefore not perfectly reproduced in our case. A much stronger absorption of light at the $D_2$ transition however is visible, which is expected given the transition dipole moment squared is larger by factor of roughly 2 compared to the $D_1$ tran-
FIG. S2. (a) Measured values of $\Delta OD_{\text{peak}}$ for various LIAD pulse peak intensities. All other parameters of the system are kept identical and comparable to the settings discussed in Fig. 3 in the manuscript. (b) Density $n(t = 2 \text{ ns})$ calculated from the fitted broadening and shift values using Eqs. (1) and (2) from the manuscript.

sition [28]. The displayed horizontal error bars in Fig. S2 show the 10% standard deviation of the LIAD peak intensity due to power fluctuations. The vertical error bar of the $\Delta OD$ is determined via error propagation from the statistical uncertainty of the photon counts $N_{\text{photon}}$, which is given by $\Delta N_{\text{photon}} = \sqrt{N_{\text{photon}}}$. The highest $\Delta OD_{\text{peak}}$ shown in Fig. S2(a) is at the limit of what can be detected with our setup due to the low number of events detected by the SPCM modules compared to the background noise of the system (dark count rate $R_{\text{dark}}$). From the datasheet provided by the manufacturer and reference measurements, we estimate at $R_{\text{dark}} \approx 1.5 \times 10^2 \text{ s}^{-1}$ with an average count rate $R_{\text{avg}} \approx 1 \times 10^6 \text{ s}^{-1}$, an overall $\Delta OD_{\text{peak}} = \ln(R_{\text{avg}}/R_{\text{dark}}) \approx 6.5$ might just be accessible. This also includes the optical depth contribution of the thermal background vapor, which was less than 0.5 for the largest cell thicknesses presented in this work ($L = 6.24 \mu$m). The observed behavior $\Delta OD_{\text{peak}}$ with $I$ could therefore be attributed to statistical limits of the SPCM in addition to the density broadening and the unknown microscopic behavior of the cell wall’s material. The latter include temporary (i.e. debris, accumulation of rubidium atoms) and permanent (i.e. cell damage) modifications for prolonged measurement cycles at one spot of the wedge-shaped cell.

The relation between a growing $\Delta OD_{\text{peak}}$ and the underlying atomic density $n_{\text{peak}}$ becomes nonlinear at high densities due to the atomic interactions and cannot be calculated directly without precise knowledge about the density broadening effect and the actual density- and velocity distribution. The actual peak density $n_{\text{peak}}$ might therefore show e.g. a linear trend with $I$, while the growth of $\Delta OD_{\text{peak}}$ appears to saturate [Fig. S2(a) data points at larger $I$]. The manuscript mentions a possible approach using steady-state derivations starting from the fitted broadening and shift. There, the presented order of magnitude of the density is calculated using Eqs. (1) and (2) from the manuscript. As the broadening and the shift lead to almost the same density values, we can calculate the average density from these two values and plot the estimated density at $t = 2 \text{ ns}$ for different LIAD peak intensities, as shown in Fig. S2(b). There, we observe an almost linear behavior between the LIAD peak intensity and the estimated density from both the $D_1$ and $D_2$ transition. The uncertainties of the estimated density result from the susceptibility fits done with EleCSUs. Note, that we apply a steady-state model to determine these density values and we have no other independent way to measure the transient density (see section V).

IV. KINEMATIC MODEL AND SIMULATION

This section describes how the kinematic model and the Monte Carlo simulation are set up to generate the optical depth map numerically [Fig. 2(b) in the manuscript].

As a first step, we pick atoms on one of the two inner cell walls ($z = 0$ or $z = L$, with the cell thickness $L$). The $x$ and $y$ position of the atoms are distributed normally, where the width of the distribution is given by the waist radius of the LIAD beam $w_{\text{LIAD}} = 13.7 \mu$m (atoms are only desorbed where the LIAD beam hits the cell). We choose a velocity distribution of the form $f(v, \varphi, \theta) = \left(4/(\sqrt{\pi}a^3)\right)v^2 \exp\left(-v^2/a^2\right)\cos(\theta)$ with the parameter $a$, adjusted to agree with the measurement.

As for the angular part of this distribution, the azimuthal angle $\varphi$ is uniformly distributed and the polar angle $\theta$ is distributed according to the $\cos(\theta)$-Knudsen law [29, 31]. The desorption time of the atoms is defined by the temporal shape of the LIAD pulse. This pulse has a shape similar to the Blackman window with a length of 1.1 ns (FWHM). The asymmetry of the measurement between the two atom clouds moving in or against the laser propagation direction (atoms with positive and negative detuning, respectively) is captured via a ratio variable $r$. This ratio $r$ is defined as the total number of atoms with negative detuning over the total number of atoms with positive detuning. After each atom traveled through the cell and hit the opposite wall, it can lead to a re-emission of another atom from the cell wall with a certain probability $p_r$ and with a new velocity ($v$: Maxwell-Boltzmann; $\varphi, \theta$: uniform). Only atoms which enter the cylindrical probe beam with a radius of $w_{\text{probe}} = 2.0 \mu$m are considered. The actual transversal Gaussian profile of the probe beam is therefore approximated by a tophat pro-
a steady-state model as an approximation. For each time step \( t \), laser detuning \( \delta \) and atom \( i \) the resulting detuning is determined as \( \Delta_{\text{atom}} = -\delta + kv_{\text{z},i} + \Delta_{\text{dd},i} \), where \( k = 2\pi/\lambda \) is the laser wave number and \( \Delta_{\text{dd}} \) is the additional dipole-dipole line shift, which depends on the local density. In addition to the natural decay rate \( \Gamma_0 \), we also include the transit broadening \( 1/\tau_{\text{tt}} \) for each individual atom to model the finite probe size effect. After including the density-dependent self-broadening \( \Gamma_{\text{self},i} \), the total broadening reads as \( \Gamma = \Gamma_0 + 1/\tau_{\text{tt},i} + \Gamma_{\text{self},i} \).

The steady-state scattering cross section is defined as [28]

\[
\sigma(\Delta_{\text{atom}}, \Gamma) = \frac{\sigma_0}{1 + 4(\Delta_{\text{atom}}/\Gamma_0)^2 + (\Gamma/\Gamma_{\text{sat}})} ,
\]  

(S8)

where \( \sigma_0 \) is the resonant scattering cross section, defined as [28]

\[
\sigma_0 = \frac{\hbar \omega_0 \Gamma_0}{2I_{\text{sat}}} .
\]

(S9)

With the additional broadening effects the scattering cross section has to be normalized with \( \Gamma_0/\Gamma \). As we are in the weak probe regime with \( I_{\text{probe}} < I_{\text{sat}} \), the scattering cross section can be written as

\[
\sigma(\Delta_{\text{atom}}, \Gamma) = \frac{\Gamma_0}{1 + 4(\Delta_{\text{atom}}/\Gamma_0)^2} \cdot \frac{\sigma_0}{(\Gamma/\Gamma_{\text{sat}})} .
\]

(S10)

To capture the dipolar dynamics of the atom-probe interaction, we use time-dependent \( \Delta_{\text{dd}} \) and \( \Gamma_{\text{self}} \) values due to the time- and \( z \)-dependent atomic density [see Eqs. (1) and (2) in the manuscript]. The scattering cross sections are accumulated and normalized for all simulated atoms within the probe region for each time step, laser detuning and \( z \) slice as \( \sigma(t, \delta, z) \). This can be understood as the average cross section \( \sigma(\delta) \) contributed by an atom found at time \( t \) at location \( z \).

The last step is the conversion of the calculated density \( n(t, z) \) and scattering cross section \( \sigma(t, \delta, z) \) to an optical depth according to the Beer-Lambert law. This correctly captures shadowing effects among atoms if the density in each \( z \) slice is low compared to its thickness (s.t. \( n\sigma L/N_c \ll 1 \) ) and can be formulated for \( z \)-dependent \( n \) and \( \sigma \). Since we only simulate the desorbed atoms without any background gas, the calculated change of the optical depth is

\[
\Delta \text{OD}(t, \delta) = \int_0^L \sigma(t, \delta, z) n(t, z) dz .
\]

(S11)

V. ELECSUS FITTING PROCEDURE

Here we show some exemplary electric susceptibility fits done with Elecsus [33]. The data points were weighted according to their inverse uncertainties shown
in Fig. S4. The error bars on the wings of the spectrum are larger due to a filter etalon in the setup, reducing the statistical uncertainty $\Delta N_{\text{photon}} = \sqrt{N_{\text{photon}}}$. We start fitting from larger times $t$, with negligible interaction, to shorter times, showing strong interaction effects, using the results from the previous fit as initial parameters for the next fit. Note that the ELECUS internal treatment of the self-broadening is manually switched off to get the full information of the broadening from the fits. We also set the Doppler temperature to $T_{\text{Doppler}} = 0 \, ^\circ\text{C}$, as we mainly want a Lorentzian profile, which is in first approximation justified to capture the self-broadening ($\Gamma_{\text{self}} > \Gamma_{\text{Doppler}}$). The free fit parameters for our ELECUS fits are the width of the Lorentzian profile ($\Gamma_{\text{buf}}$, normally used for a buffer gas broadening), the line shift and the temperature of the cell $T_{\text{cell}}$, which is used to calculate a temperature-dependent atomic density. Note, that this intrinsic temperature-dependent density in ELECUS is not a useful quantity in our anisotropic system and therefore not considered in this work. In our system the velocity of the atoms has a certain direction, where a possible velocity distribution is discussed in our kinematic model (see also Section IV).

The ELECUS software calculates the 1σ standard error of the fitted variables. These standard errors are plotted as vertical error bars in Fig. 4 in the manuscript and in Fig. S5. There are no horizontal error bars shown in both these figures, as they originate from histogram binning. In this sense, the horizontal precision is limited by the bin width which is motivated by the total temporal precision $\Delta t_{\text{total}} = 850 \, \text{ps}$ of each captured event. This $\Delta t_{\text{total}}$ is the sum of the LIAD pulse-to-pulse jitter and the time resolution of the SPCM, which limit the bandwidth of the measurement. The goodness of each fit is evaluated by calculating the normalized root-mean-square deviation $\text{NRMSD}$ as given in the manuscript, which is calculated from the root-mean-square deviation $\text{RMSD} = N^{-1} \sqrt{\sum_{i=1}^{N} (T_i - \hat{T_i})^2}$ between the measured transmission points $T_i$ and the fitted transmission value $\hat{T}_i$. These are then normalized using the minimum and maximum transmission values $T_{\text{min}} = 0, T_{\text{max}} = 1$ as $\text{NRMSD} = \text{RMSD}/(T_{\text{max}} - T_{\text{min}})$. The obtained values are similar for both probe transitions and typically at $\text{NRMSD} \approx 5\%$.

The electric susceptibility model is a steady-state solution of the atom-light interaction. As mentioned in the manuscript, we cannot properly fit the data points in the...
The ratio between the line shifts can be properly recovered from the fits for $t < 7 \text{ ns}$ even if the wings are not fully captured, as the the dipole-dipole shift is smaller than the self-broadening and can be measured as an absolute offset instead of a line width. For very large times, corresponding to low densities, the ratio between the vanishing shifts on the $D_1$ and $D_2$ transition will be very susceptible to any residual offset. In this region, which occurs at different times for self-broadening and line shift due to their different absolute values, we observe varying and unstable behavior. We suspect that a combination of all these effects causes deviations from the theoretical ratios between the $D_1$ and $D_2$ transition as reported in Fig. 4 in the manuscript.

While the self-broadening is directly visible in Fig. S4 for exemplary times, we also show the fitted line shift by drawing the frequencies of the two ground state hyperfine splittings of the two rubidium isotopes, to emphasize the changing line shift over time. The figure also highlights that artifacts due to a clipped laser detuning range would impact the $D_1$ transition less (or at different times) than the $D_2$ transition. This idea is supported by the reduced variation in the $D_2/D_1$ ratio in Fig. S5(a), which indeed has the lowest overall density and broadening.

To underline our observation of the transient density-dependent dipolar interactions, we show the time evolution of the self-broadening for two additional LIAD intensities in Fig. S5. With increasing intensity, the density is also increasing, leading to a larger self-broadening. The ratio of $D_2/D_1$ shows a similar behavior for all intensities.

VI. PULSED LIAD PROBED AT 420 nm

The experiments in the manuscript were performed using probe lasers at the $D_1$ and $D_2$ ground state transitions, with relatively large transition dipole moments.

FIG. S5. Self-broadening of the $D_1$ (yellow) and $D_2$ (purple) transition over time for three different LIAD intensities $I$. Additionally, the ratio $D_2/D_1$ (green) is plotted, which is approaching the theoretical steady-state ratio (black triangle). For high intensities, e.g. in (c), the broadening of the $D_2$ transition is overestimated during the fit (see also Fig. S4), which is one reason for the deviation of the ratio from the theoretical value in the first $\approx 10 \text{ ns}$.

first 2 ns. Initially the broadening is large and the signals’ wings are not properly captured with $\approx 2\pi \times 16 \text{ GHz}$ scan range in our experiment. Also the LIAD pulse, which is present until $t \approx 1.5 \text{ ns}$, distorts the measured data. There are several effects which could systematically distort the quality of the fit results or render the model unsuitable for later times $t$ (ordered by the hypothesized contribution from strongest to lowest according to the authors): A non-isotropic velocity distribution as produced by the LIAD pulse, geometry-dependent effects like the collective Lamb shift, mathematical artifacts due to a clipped detuning range, asymmetries from different relative hyperfine transition strengths and dipole moments, asymmetric line shapes caused by any other effect (e.g. surface potentials), multi-particle interactions, differences in the experimental configuration between both transitions, and temporally transient dipolar effects.

The ratio between the line shifts can be properly recovered from the fits for $t < 7 \text{ ns}$ even if the wings are not

FIG. S6. Measured $\Delta \text{OD}$ map on the $5S_{1/2} \rightarrow 6P_{1/2}$ transition at a cell thickness of 2.34(4) µm and a reservoir temperature of $T_{\text{res}} \approx 230^\circ \text{C}$. The four green markers indicate the ground state hyperfine splitting of the two isotopes of rubidium, respectively. Note, that the image has been filtered using a Gaussian filter to reduce the overall noise in the data. The intensity $I$ is the peak intensity of the LIAD pulse.
This, however, induces limits considering the accessible density regimes before broadening effects become larger than the captured frequency range as discussed above. It can be overcome by probing on a transition with a lower transition dipole moment.

We therefore modified our measurement setup to include a 421.6 nm laser probing the $5S_{1/2} \rightarrow 6P_{1/2}$ transition, where the squared transition dipole moments are reduced by roughly two orders of magnitude [30]. The measured data for total integration times similar to the previously discussed cases exhibits a much noisier signal and lower $\Delta$OD, which is expected due to the much weaker transition dipole moment. Consequently, the resulting map shown in Fig. S6 could only be captured for relatively strong LIAD pulses, a thicker part of the cell, and a higher reservoir and cell temperature. Important features discussed in the manuscript are reproduced. Both signals representing atoms moving towards and away from the laser are visible and exhibit the same asymmetry towards atoms moving in propagation direction of the LIAD laser. We were not able to measure a line shift or broadening which is attributed to the significantly weaker transition dipole moment. This further supports the conclusion that the observed effects in the manuscript indeed have a dipolar origin.

F.C., M.M., and F.M. contributed equally to this work.