Observation of glassy dynamics in a disordered quantum spin system

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Understanding the dynamics of strongly interacting disordered quantum systems is one of the most challenging problems in modern science, due to features such as the breakdown of thermalization and the emergence of glassy phases of matter. We report on the discovery of anomalous slow relaxation dynamics in an isolated quantum spin system realized by a frozen gas of Rydberg atoms. The total magnetization is found to relax sub-exponentially analogous to classical glassy dynamics yet being governed by the build-up of non-classical correlations. Experimentally as well as in semi-classical simulations, we find the evolution towards a randomized state to be independent of the strength of disorder up to a critical value. This suggests a unifying description of relaxation dynamics in disordered isolated quantum systems, analogous to the generalization of statistical mechanics to out-of-equilibrium physics in classical spin glasses.

The far from equilibrium behavior of isolated quantum spin Hamiltonians with disorder is still relatively poorly understood. This is in part due to the fact that most condensed matter systems are not fully isolated from their environment, and hence always relax to the thermal equilibrium that is imposed by the bath [1]. However, disorder and frustration in open many-body quantum systems lead to a breakdown of thermalization on experimentally accessible timescales [2]. Instead, such systems show anomalously slow relaxation of macroscopic observables in a characteristically non-exponential way which seems to be a unifying feature encountered in e.g., spin glasses [3], quasi crystals [4], organic superconductors [5], atoms in optical lattices [6] or diamond color centers [7, 8]. In contrast, for isolated quantum systems it is an open question which fundamental processes govern the dynamics towards equilibrium. Recently it was conjectured that even isolated quantum systems evolving according to fully unitary dynamics also generically relax to a state of local thermal equilibrium according to the eigenstate formalization hypothesis (ETH) [9], possibly via long-lived pre-thermal states [10–12]. Here, we encode a quantum pseudo-spin-1/2 in two electronically excited (Rydberg) states of each atom comprising a frozen atomic gas [13]. Spin-spin interactions arise from the state-dependent van der Waals interactions between Rydberg states, while disorder originates from the random positions of each atom in the gas which gives rise to distance dependent couplings. Thus, our system represents a quantum simulator for unitary spin dynamics far-from-equilibrium [14–19].

To give a qualitative picture of the quantum dynamics in our system, we consider a small ensemble of 12 spin-1/2 particles randomly positioned in space and all initialized in the $|↓⟩$ state, corresponding to an initial magnetization $S_z = 1/2$. The experimental protocol is depicted in Figure 1A. The unitary dynamical evolution of the system is governed by the Heisenberg XXZ-Hamiltonian in the absence of magnetic fields (in units where $\hbar = 1$),

$$H_{XXZ} = \frac{1}{2} \sum_{i,j} J_{ij}(S_x^{(i)}S_x^{(j)} + S_y^{(i)}S_y^{(j)} + \delta S_z^{(i)}S_z^{(j)}),$$

(1)

where $\delta$ is the anisotropy parameter and $J_{ij}$ are the interaction couplings between the spins $i$ and $j$ (see Fig. 2B). We focus on spin-spin interactions that decay as a power law $J_{ij} = C_0/r_{ij}^{\gamma}$ with the inter-particle distance $r_{ij}$ (for the derivation of the Hamiltonian, see [20]). The results of a full quantum mechanical simulation is shown in Fig. 1C. Due to the spatial disorder, spin-spin interactions give rise to complex many-body dynamics on strongly varying timescales in a single disorder realization (see grey curve in Fig. 1C).

The loss of magnetization is accompanied by a
FIG. 1. Relaxation dynamics in a disordered quantum spin system. (A) Experimental protocol for initialization and readout of the many-body spin system composed of Rydberg atoms. Spin $\uparrow$ and $\downarrow$ correspond to two different Rydberg states. (B) Level diagram of a pair of spin-1 particles. The interactions $J_{ij}$ and $\delta J_{ij}$ entering the Hamiltonian (1) arise from the interactions between Rydberg atoms [20]. (C) Exact simulation of 12 spins interacting via a Heisenberg XXZ Hamiltonian. The plot shows the magnetization for a single realization (grey curve) and the disorder average over 1000 realizations (black curve) which relaxes as function of time given units of the median interaction strength $J_{\text{med}}$. The dotted line indicates the mean-field prediction that does not relax. The dashed black line is a guide to the eye highlighting the logarithmic timescale of the dynamics. The microscopic expectation values of $\langle S_i^{(1)} \rangle$, $\langle S_i^{(y)} \rangle$ and $\langle S_i^{(z)} \rangle$ for each spin are plotted at three different time steps on the Bloch sphere. The reduction of the expectation values (magnetization) is a consequence of the spreading of entanglement (visualized by the blue bonds between spins).

build-up of entanglement between spins witnessed by a reduction of the local purity of each spin [20]. This behaviour is in stark contrast to an effectively classical, mean-field prediction for this Hamiltonian (dotted line in Fig. 1c), which assumes each spin to evolve in the average field generated by all other spins thus neglecting quantum correlations. In fact, the initial fully magnetized state is an eigenstate of the mean-field Hamiltonian resulting in a total absence of relaxation. After ensemble and disorder averaging the magnetization approaches a fully randomized state with $\langle S_i^{(z)} \rangle = 0$ (see black curve in Fig. 1C), consistent with the ETH prediction. However this relaxation occurs very slowly compared to the timescales associated with spin-spin interactions, and appears to follow an almost logarithmic dependence (dashed line in Fig. 1C). It remains an open question whether such glassy dynamics can actually be observed in fully isolated quantum many-body systems and to what extent it shares common features with classical spin glasses.

We address this question with our experiments that are performed using a gas of ultracold rubidium atoms held in an optical trap and then prepared in a superposition of two different Rydberg states: $|\downarrow\rangle = |48s\rangle$ and $|\uparrow\rangle = |49s\rangle$. The experimental procedure (Fig. 1A) starts with a laser pulse of variable duration that brings the atomic ground state to the $|\downarrow\rangle$ state with an approximately three-dimensional Gaussian density distribution consisting of between 400 and 1200 spins. Next a microwave $\pi/2$ pulse coupling $|\downarrow\rangle \leftrightarrow |\uparrow\rangle$ rotates all spins to the fully-magnetized state $|\rightarrow\rangle_{\otimes N}$ with all spins pointing along the $x$-direction on the Bloch sphere. This state has been chosen since it would not undergo any dynamics according to the classical equations of motion and is thus ideally suited to elucidating the role of quantum effects on the dynamics. On the timescale of our observations, atomic motion can be neglected (frozen gas limit). The time evolution is governed by the Hamiltonian (1), with $C_{\text{dip}}/2\pi = 64 \text{ GHz} \mu\text{m}^6$ characterizing the strength of the interactions which arise from dipole-dipole couplings between pairs of atoms. After a time evolution $t$, a second microwave $\pi/2$ pulse with adjustable phase is applied to rotate the equatorial magnetization components to the detection basis (Fig. 1c). In this way we effectively read out the $\langle S_x \rangle$ and $\langle S_y \rangle$ magnetizations from population measurements of the two spin states $|\downarrow\rangle$ and $|\uparrow\rangle$ using electric field ionization [20].

Fig. 2 shows the experimentally observed relaxation of the magnetization using tomographic spin-resolved readout of the $\langle S_x \rangle$ and $\langle S_y \rangle$ magnetizations. Starting from the almost fully magnetized state $\langle S_z \rangle = 1/2$ we observe that the magnetization decays towards the unmagnetized state within $\approx 10 \mu s$. This is much shorter than the independently measured single-spin coherence time of $\gtrsim 100 \mu s$ ([20]), but still orders of magnitude
of 120 realizations of the experiment at a peak spin density. Error bars are determined from the two magnetization components \(\langle S_x \rangle \) and \(\langle S_y \rangle \) from a tomographic spin readout. The observed dynamics are clearly inconsistent with the mean-field prediction including imperfect initial state preparation (dotted line). The solid lines are dTWA predictions without free parameters. The shaded area indicates the systematic uncertainty of the measured density. The dashed line depicts a fit of the data with a stretched exponential function (dashed line in Fig. 2) yielding an exponent \(\beta = 0.32(2)\). This value clearly rules out a pure exponential decay, i.e. \(\beta = 1\) that could be expected on the basis of single-particle dephasing. It is also inconsistent with an exponent of \(1/2\) that is expected for an Ising model [23, 24], a fluctuator model for open quantum systems [7, 8] or an averaging of local exponential decays weighted by the inhomogeneous Gaussian spin distribution [20]. Our observation therefore indicates a close similarity between classical glassy dynamics and quantum glassy dynamics in the form of subexponential relaxation of the macroscopic magnetization [25].

To further investigate how the slow relaxation and the characteristic exponent depends on microscopic details, we control the degree of spatial disorder by taking advantage of the Rydberg blockade effect during state preparation [26, 27]. During laser excitation the strong van der Waals interactions between Rydberg states prevents two spins to be prepared at distances smaller than the Rydberg blockade radius \(R_{bl}\). The degree of disorder is thus controlled by the ratio \(a/R_{bl}\) where \(a\) is the Wigner-Seitz radius for the spin distribution \(a = (4\pi \rho_S^0/3)^{-1/3}\) (Fig. 3a). For \(a/R_{bl} \rightarrow 0\) the blockade effect has little influence and the spins are randomly distributed, whereas the limit \(a/R_{bl} \rightarrow 1\) corresponds to a fully ordered configuration. In between (when \(R_{bl}\) becomes comparable to \(a\), the short distance cutoff imposed by the Rydberg blockade effect effectively reduces the strength of the disorder compared to fully uncorrelated random spin positions. In the experiment we can tune the disorder strength by changing the peak spin density \(\rho_S^0\) and thus the mean number of spins per blockade sphere \((a/R_{bl})^{-3}\) from \(0.20(5)\) to \(0.7(2)\) (two-dimensional representations of corresponding distributions are depicted in Fig. 3a). Remarkably, we find the stretched exponent \(\beta\) to be effectively independent over this range (inset in Figure 3b). Furthermore, after rescaling the time axis by the characteristic energy scale \(C_0/a^6\), the time-dependent data collapse onto a single line (Fig. 3b). This suggests that the dynamics are universal, i.e. independent of the microscopic details of the system, an observation that will be explored further in a theoretical model.

To simulate the three-dimensional Rydberg gas on
A classical computer, we model the system by an ensemble of spin-1/2 particles with interactions described by the Heisenberg XXZ-Hamiltonian. Due to the large number of spins in the experiment, the full simulation of the unitary dynamics of the Hamiltonian Eq. (1) is not possible. Instead, quantum effects can be partially taken into account by applying the semiclassical discrete Truncated Wigner Approximation (dTWA, [20]) [28, 29] which has recently been shown to describe the dynamics of Rydberg interacting spin systems very well [13]. To model the present experiments, all relevant parameters for the simulation are determined through independent measurements, such as the spatial density distribution, total number of spins and the microwave coupling strength $\Omega$ used in the preparation and readout stage. The initial spin distribution is obtained from a random excitation model of the Rydberg atoms, including a cutoff distance to account for the blockade effect [20].

The numerical simulations describe remarkably well the glassy dynamics, further confirming the validity of the dTWA approximation for treating the dynamics of disordered quantum systems [13]. This ability to reproduce the experimental observations using dTWA simulation also suggests a possible explanation in analogy to classical spin glasses: quantum, instead of thermal fluctuations, allow the system to explore a rugged energy landscape leading to a hierarchical separation of time-scales [25, 30], resulting in stretched exponential relaxation. Theoretical modeling using the dTWA allows to further test the universal behaviour and exclude possible effects of the inhomogeneous spin density resulting from the optical trap. Rescaled spin dynamics in a uniform particle distribution are shown in Fig. 4. For early times the leading order quadratic Hamiltonian evolution is clearly visible. For times beyond the perturbative regime we find that the rescaled data collapse on a single stretched exponential curve characteristic of glassy quantum dynamics.

Investigating the dTWA simulations for even higher densities than accessible in the experiment (corresponding to more correlated spatial configurations of the atoms) shows that the stretching exponent $\beta$ is sensitive to the strength of the disorder above a threshold. The stretched exponent $\beta > 1$ indicates a breakdown of the glassy dynamics for more ordered systems. To quantify this we use the Kullback-Leibler divergence $D_{KL}$ of the distribution of nearest-neighbour interaction strength with respect to a completely disordered one. By definition, it is zero for uncorrelated distributions and diverges for atoms on a regular lattice. The inset of Fig. 4 shows the resulting exponent as function of $D_{KL}$. For disorder strengths $D_{KL} \lesssim 1$, we find a universal stretched exponent of $\beta = 0.36$ close to the experimental value. In this regime the cutoff is much larger than the median interaction strength and the functional form of the spin distribution function is mainly modified at strong interactions. This can explain why only the early time dynamics is changed.
and glassy dynamics is observed. Still it is remarkable that changes in the overall distribution function do not affect the exponent as could be derived by a simple model involving averaging over exponential decays with rates given by a local mean interaction strength [20]. The fact that the exponent is constant over a wide range of $\mathcal{D}_{KL}$, provides evidence that glassy dynamics is not significantly influenced by the inhomogeneous density or the strength of disorder over the range of parameters experimentally explored. Therefore, applying dTWA for the actual inhomogeneous spin distribution results in a density independent stretched exponent $\beta$ which is in excellent agreement with the measurements as shown by the inset in Fig. 3. In this work we observed glassy dynamics reminiscent of the subexponential relaxation that is known from classical spin glasses. While the latter is driven by thermal fluctuations, the dynamics of the disordered isolated quantum system is governed by quantum fluctuations and spreading of entanglement going beyond mean-field approximations. Remarkably, the stretching exponent $\beta$ appears to be a universal parameter below a certain disorder strength, as confirmed by both experiment and semi-classical simulations. In the experiment, disorder is changed by controlling of the density of spins, which actually shifts the upper cutoff scale in the distribution of interaction strengths due the excitation blockade effect. Thus, the long-time evolution is insensitive to the details of the system parameters on high energy scales which hints at a low-energy effective theory valid at long times. In addition, we observe that in this regime the dynamics of the magnetization is well described by semi-classical truncated Wigner simulations suggesting that the significance of quantum interference effects decreases as the system approaches its equilibrium state. This is in line with the finding that the long-time dynamics of generic thermalizing quantum many-body systems simplifies also in the sense that states can be represented efficiently due to their limited entanglement [31]. We take the observed independence of high-energy details and applicability of semi-classical description as strong hints that simplified effective descriptions of the dynamics of quantum many-body systems at long times, i.e. close to thermal equilibrium, exist. The analogy to classical spin glasses where the fluctuation-dissipation theorem has been found to hold in the aging regime [32] and the spin-glass transition shows similarities to thermal phase transitions [33] highlights numerous opportunities to find an effective description of glassy dynamics in the quantum regime.

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![Figure 4](image-url)
is detuned from the intermediate state $|48P_{3/2}\rangle$ by 170 MHz, far enough to guarantee a residual population on this state smaller than 4%. The atoms can therefore be considered as two-level systems and described by a pseudo-spin degree of freedom $S^{(i)}$. The microwave field acts on the spins as an external field and is described by the Hamiltonian

$$H_{\text{ext}} = \sum_i \Omega \cos \phi S_x^{(i)} + \Omega \sin \phi S_y^{(i)} + \Delta S_z^{(i)},$$

with $\Omega$ the Rabi frequency, $\Delta$ the detuning and $\phi$ the phase of the field. To vary these three parameters at the 10 ns time scale, we use frequency up-conversion with a radio-frequency field at 400 MHz.

The preparation and readout $\pi/2$ pulses are performed at an effective Rabi frequency $\Omega/(2\pi) = 3.00(1)$ MHz and a detuning $\Delta/(2\pi) = 0$. The Rabi frequency is extracted from the period of Rabi oscillations between the two spin states. By convention, the phase $\phi$ of the preparation pulse is set to 0, but it can be tuned for the readout pulse to measure the magnetization $\langle S_\phi \rangle$ in any direction of the equatorial plane:

$$\langle S_\phi \rangle = \cos \phi \langle S_x \rangle + \sin \phi \langle S_y \rangle.$$  

Uncertainties in the duration and amplitude of the pulses as well as interaction effects lead to imperfect initial spin state. Based on simulations, we estimate the fidelity to be higher than 96%.

**Determination of the magnetization**

The magnetization is extracted from population measurements of the Rydberg states using electric field ionization [35]. At the end of the sequence, a strong electric field of 100 V cm$^{-1}$ is switched on and the resulting ions are guided towards a multichannel plate detector. To calibrate the detection efficiency, we combine ionization measurements and depletion imaging. We deduce a detection efficiency $\eta = 0.173 \pm 0.004$ from four different calibration curves.

At time $t$ we access the magnetization $\langle S_\phi \rangle$ by counting after the readout pulse both the population of the $|\uparrow\rangle$ state $N_\uparrow(\phi)$, and the total spin number $N_{\downarrow+\uparrow}$, according to

$$\langle S_\phi \rangle = \frac{N_\uparrow(\phi) - N_\downarrow(\phi)}{2N_{\downarrow+\uparrow}} = \frac{N_{\uparrow}(\phi)}{N_{\downarrow+\uparrow}} - \frac{1}{2}.$$  

**Spin manipulation and Ramsey sequences**

A two-photon microwave field is then used to couple the $|\downarrow\rangle$ state to the $|\uparrow\rangle = |48S_{1/2}, m_j = +1/2\rangle$ Rydberg state. The single photon frequency $\nu = 35.2$ GHz is detuned from the intermediate state $|48P_{3/2}\rangle$ by 170 MHz, far enough to guarantee a residual population on this state smaller than 4%. The atoms can therefore be considered as two-level systems and described by a pseudo-spin degree of freedom $S^{(i)}$. The microwave field acts on the spins as an external field and is described by the Hamiltonian

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Since the ionization is not state-selective, \( N_\uparrow(\phi) \) is inferred by counting the spin number after depopulating the \( |\downarrow\rangle \) state. It is performed by optically coupling the \( |\downarrow\rangle \) state to the short-lived intermediate state \( |e\rangle \) during 1.5 \( \mu s \).

Due to the finite lifetime of the spin states and microwave transfer, auxiliary Rydberg states might also be populated. This residual population leads to an offset in the measured ion signal, a number \( N_a \) of those atoms being energetically above the ionization threshold (see Fig. 5). As a consequence, what we measure instead of \( N_\uparrow(\phi) \) and \( N_{\downarrow+\uparrow} \) are two quantities \( M_\uparrow(\phi) \) and \( M_{\downarrow+\uparrow} \) given by

\[
M_\uparrow(\phi) = N_\uparrow(\phi) + N_a, \quad (7)
\]

\[
M_{\downarrow+\uparrow} = N_{\downarrow+\uparrow} + N_a. \quad (8)
\]

The measured quantity \( M_\uparrow \) is a sinusoidal function of \( \phi \), centred around its mean value

\[
\overline{M}_\uparrow = \frac{N_{\downarrow+\uparrow}}{2} + N_a. \quad (9)
\]

We determine from a sinusoidal fit the values \( M_\uparrow(\phi) \) and \( \overline{M}_\uparrow \) and thus compute the magnetization \( S_\phi \) using eqs. (6) to (9). The amplitude \( A \) of the sinusoidal fit, normalized by \( N_{\downarrow+\uparrow} \), corresponds to the magnetization in the \( xy \)-plane.

\[
\frac{\overline{M}_\uparrow}{N_{\downarrow+\uparrow}} = A.
\]

Following this procedure, we deduce that the number of atoms in the auxiliary states \( N_a \) increases linearly in time with a rate of 7 kHz, consistent with the blackbody decay of the spin states toward Rydberg states above the ionization threshold.

**Measurement of the single spin phase coherence time**

To check the assumption of unitary Hamiltonian evolution with negligible effects of decoherence, we have performed a Ramsey measurement at low spin densities where interactions are negligible (Fig. 6, left panel). The full contrast oscillation shows that the single spin coherence time is as long as the depopulation time of the Rydberg states through spontaneous decay and redistribution by black-body radiation (113 \( \mu s \) and 121 \( \mu s \) for the chosen Rydberg states [36]) and that single particle dephasing can be neglected on our experimental time scales ranging up to 10 \( \mu s \). At higher densities (Fig. 6, central and right panel), however, we observe a decay of the oscillation contrast which increases with increasing density, which indicates that this decay is caused by the spin-spin interactions.

\[
\frac{\overline{M}_\uparrow}{N_{\downarrow+\uparrow}} = A.
\]

**Spatial spin distribution and disorder strength**

To model the experimental 3D spin distribution, we simulate the Rydberg excitation dynamics in a cloud of ground state atoms with Gaussian density distribution (measured radii at \( 1/e^2 \): \( \sigma_x = 203(3) \) \( \mu m \), \( \sigma_y = \sigma_z = 35(1) \) \( \mu m \)). We excite each atom randomly one after the other with a certain probability if it is not within the blockade sphere of an already excited atom. The excitation probability takes into account the \( \sqrt{N_b} \) enhancement due to Rydberg interactions, with \( N_b \) the...
number of atoms within a blockade sphere. It also consider the profile of the laser excitation, characterized by a Gaussian distribution of the two-photon Rabi frequency with measured radius $\sigma = 70.6(3)$ $\mu m$ $(1/e^2)$. Its amplitude is adjusted such that the total number of excited atoms equals the one measured by field ionization.

For simulations of a homogeneous system, the spins are randomly distributed in a uniform box taking into account a blockade radius of $5 \mu m$ until the desired density is reached. In the limit of no blockade effect, the nearest-neighbour distribution would be given by [37]

$$h(r) = \frac{3}{a} (r/a)^2 \exp\left(-\frac{(r/a)^3}{6}\right),$$  \hspace{1cm} (10)

yielding the distribution of coupling strengths $h(J) = h(r) \frac{8}{27}$. Instead, the blockade effect modifies the nearest-neighbour distribution, resulting in a different coupling distribution $g(J)$ (see Distribution of interaction strengths). We quantify the disorder strength of the spin system with the Kullback-Leibler divergence [38]

$$D_{KL}(g \parallel h) = \int g(J) \log \left( \frac{g(J)}{h(J)} \right) \, dJ,$$  \hspace{1cm} (11)

i.e. the amount of information that is gained by updating from the distribution $h(J)$.

When investigating the universal character of the spin dynamics for homogeneous system, we have concluded that the typical energy scale should be determined by $C_6/a^6$ (see Fig. 4). The effective distance $\tilde{a}$ is defined as the median of the non-Gaussian distribution $\{\tilde{r}_i\}$ with $\tilde{r}_i^{-6} = \sum_j r_{ij}^{-6}$. The energy scale $C_6/a^6$ is thus the median of the mean-field energies $C_{6}/r_{ij}^6$. This typical energy is well suited to rescale the dynamics since it agrees precisely with the fitted relaxation rate $\gamma_J$ for the explored densities (see Fig. 7).

For low density, the effective distance $\tilde{a}$ almost coincides with the Wigner-Seitz radius $a$ (see Fig. 7). It thus justifies the analysis performed in Fig. 3, the experimental data exploring a range of low to intermediate densities where deviations of $\tilde{a}$ to $a$ can be neglected.

**Calculation of Rydberg interactions and Spin model**

In order to describe the interaction between two Rydberg excitations, the Hamiltonian is expanded in multipoles. This is well justified, as the minimal distance between the Rydberg atoms that is determined by the blockade radius $R_{bl}$ is much larger than the LeRoy radius $R_{LR}$ that describes the typical spread of the electron wave function. The leading order term of this expansion is the dipole-dipole interaction Hamiltonian

$$\hat{H}_{DDI} = \frac{\hat{d}_i \cdot \hat{d}_j - 3 (\hat{d}_i \cdot e_r) (\hat{d}_j \cdot e_r)}{R^3},$$  \hspace{1cm} (12)

that couples Rydberg atoms with different angular moment quantum number $l$. For dipolar forbidden transitions, the second order term in perturbation theory needs to be calculated giving rise to the van der Waals Hamiltonian

$$H_{vdW} = -\frac{1}{\hbar} \sum_m \frac{H_{DDI}[m]\langle m | H_{DDI} | \Delta_F \delta (\omega_j m + \omega_m)$$  \hspace{1cm} (13)

where the Foerster defect $\Delta_F = E_m - E_i$ is the energy difference between the intermediate and initial state. Aiming for a simpler notation, the two different Rydberg states can be identified as spin states $|\uparrow\rangle$ and $|\downarrow\rangle$. In the pair state basis $\{|\uparrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$, the total Hamiltonian describing the interaction between two atoms $i$ and $j$ can be written in matrix form as

$$\hat{H}^{tot}_{i,j} = \begin{pmatrix} E_{\uparrow\uparrow} & 0 & 0 & 0 \\ 0 & E_{\uparrow\downarrow} \frac{\Delta_F}{2} & 0 & 0 \\ 0 & \frac{\Delta_F}{2} & E_{\downarrow\uparrow} & 0 \\ 0 & 0 & 0 & E_{\downarrow\downarrow} \end{pmatrix}.$$  \hspace{1cm} (14)
with the matrix elements $E_{\uparrow\uparrow} = \langle \uparrow\uparrow | H_{\text{vdW}} | \uparrow\uparrow \rangle$, $E_{\downarrow\uparrow} = \langle \downarrow\uparrow | H_{\text{vdW}} | \uparrow\downarrow \rangle$, $E_{\downarrow\downarrow} = \langle \downarrow\downarrow | H_{\text{vdW}} | \downarrow\downarrow \rangle$ and $J_{\text{ex}} = \langle \uparrow\downarrow | H_{\text{vdW}} | \downarrow\uparrow \rangle$. This Hamiltonian can be identified as the Heisenberg XXZ Hamiltonian

$$H_{\text{XXZ}} = \frac{1}{2} \sum_{i,j} J_{ij} (S_x^{(i)} S_x^{(j)} + S_y^{(i)} S_y^{(j)} + \delta S_z^{(i)} S_z^{(j)}) + \sum_i \Delta_{\text{vdW}} S_z(i)$$

where $J_{ij} = 2J_{\text{ex}}$, $\delta = (E_{\downarrow\downarrow} + E_{\uparrow\uparrow} - 2E_{\downarrow\uparrow})/J_{ij}$ and $\Delta_{\text{vdW}} = (E_{\downarrow\downarrow} - E_{\uparrow\uparrow})/2$. The additional single-spin detuning $\Delta_{\text{vdW}}$ is an order of magnitude smaller than the interaction strength $J_{ij}$ and thus negligible.

The matrix elements $E_{\uparrow\uparrow}$, $E_{\downarrow\uparrow}$, $E_{\downarrow\downarrow}$ and $J_{\text{ex}}$ were calculated using the python module ARC [36]. For the Rydberg states $|48S_z, +\frac{1}{2}\rangle$ and $|49S_z, +\frac{1}{2}\rangle$ this yields the interaction strength $J_{ij} = C_6/r_{ij}$ with $C_6/(2\pi) = 59\text{ GHz}\cdot\mu\text{m}^6$ and $\delta = -0.73$.

**Distribution of interaction strengths**

When the spin density increases, the spatial distribution of spins becomes more and more affected by the blockade effect, leading to increasing order. For a uniform distribution of Rydberg atoms with fixed blockade radius, the Kullback-Leibler divergence increases approximately linear with density (see inset in Fig. 8).

As an illustration, we plot in Fig. 8 the distribution of nearest-neighbour interaction strengths. For the minimal density in the experiment, the probability distribution (orange solid line) is very similar to the completely random case (orange dashed line). For the maximal density of the experiment, the blockade effect introduces a cut-off for large interaction strengths that changes significantly the probability distribution (green solid line) compared to the random case (green dashed line).

**Theoretical models**

To compare the experiment to the mean-field prediction, we solve the classical equations of motion that are obtained from the classical Hamiltonian function [29]

$$H_C = \frac{1}{2} \sum_{i,j} J_{ij} (s_x^{(i)} s_x^{(j)} + s_y^{(i)} s_y^{(j)} + \delta s_z^{(i)} s_z^{(j)})$$

$$s_x^{(j)} = \{ s_x^{(j)}, H_C \}. \quad \text{(15)}$$

Here, $s^{(i)} = (s_x^{(i)}, s_y^{(i)}, s_z^{(i)})$ are classical spins and $\{ \ldots \}$ denotes the Poisson bracket. For a perfect initial state where all spins are aligned in $x$-direction, mean-field theory does not predict any dynamics. However, the interactions present during the first $\pi/2$-pulse of the Ramsey protocol induce small fluctuations in the initial state. These lead to the dynamics shown by the dotted line in Fig. 2. For the relevant time-scale of the experiment, these dynamics are negligible.

For the considered dynamics, the initial state is an eigenstate of the mean-field Hamiltonian. The relaxation is thus triggered by the initial quantum fluctuations, meaning that mean-field approaches fails in this case. Instead, we use a Truncated Wigner Approximation (TWA), which still performs classical evolution of the spins but includes the initial quantum fluctuations into statistical ensembles of the initial state [28, 29]. This approach has been successfully applied to spin systems in recent work [13]. Imperfections of the preparation and readout are taken into account by simulating the whole sequence, including those two stages.
We also compared TWA with an approximate quantum mechanical model, the so-called Moving Average Cluster Expansion [39] which qualitatively give similar results (see Fig. 9).

![Graph of magnetization over time](image)

**FIG. 9.** Comparison between dTWA and MACE. Both, MACE and dTWA perform well to describe the experimental data at late times, at intermediate times (between 1 and 3 µs), MACE predicts faster depolarisation dynamics.

Quantification of slow dynamics by a stretched exponential

A phenomenological approach to describe slow dynamics in disordered systems is a fit of the magnetization with a stretched exponential

\[
S_x(t) = \frac{1}{2} \exp(-\gamma_J t)^\beta
\]

with relaxation rate \(\gamma_J\) and stretched exponent \(\beta\). This was already proposed by Kohlrausch in 1847 [40], a review on the stretched exponent in numerical simulations and in experimental data of various materials can be found in [21].

For \(\beta = 1\) the stretched exponential describes an exponential decay. In the limit \(\beta \to 0\), the stretched exponential approaches the logarithmic decay which can be seen by performing a Taylor expansion at small \(\beta\)

\[
\exp(-t/\tau)^\beta = \frac{1}{e} - \frac{\beta \log(\frac{1}{2})}{e} + \mathcal{O}(\beta^3).
\]

Therefore, we exclude the very early dynamics from the fit where \(t < 1/J_{\text{max}}\) (see Fig. 4).

**FIG. 10.** Relaxation of the magnetization in a uniform system for different \(D_{KL}\). Time evolution of the magnetization and fits of the stretched exponential for the same simulations as in Figure 4. The early time dynamics is excluded up to \(t = 1/J_{\text{max}}\).

Slow dynamics inconsistent with an average over exponential decays

It is an open question how to explain the slow dynamics and especially the seemingly universal stretched exponent \(\beta\) for strongly disordered systems. Stretched exponential dynamics are typically explained by an average over exponential decays [21], however in our isolated system it is not trivial to identify those decay channels. A first guess where the decay rates
Averaged exponential decays. (A) The average over exponential decays with rates given by $C_6/\bar{r}_i^6$ (solid line) and a fit with a stretched exponential decay (dotted line).

(B) Comparison of the stretched exponent $\beta$ for simulations with dTWA and the average over exponential decays.

are given by $C_6/\bar{r}_i^6$, reproduces stretched exponential functions for disordered spin distributions and an increase of $\beta$ for less disordered systems (see Fig. 11).

However, this approach cannot explain the existence of the regime with constant $\beta$ for $D_{KL} \lesssim 1$. In the infinitely disordered limit, the sum over exponential decays predicts a stretched exponent of $\beta = 0.5$. This value of $\beta$ agrees with analytic results obtained by integrating over a Gaussian distribution of decay rates. The discrepancy to the numerical and experimental value is similar to the one observed by Fischer et al. [22] and might be explained by higher order correlations propagating in the system.

**Entanglement**

After the discussion of the slow relaxation dynamics, we want to stress out the significance of quantum fluctuations for the dynamics by providing a link between the relaxation of the magnetization and the build-up of entanglement between an individual spin and the remaining system. This is remarkable, because typically, the entanglement is difficult to measure and the full density matrix $\rho_i$ of each spin needs to be known. In order to partly overcome this problem, we will exploit the fact, that the spins show no classical dynamics (see Figure 2).

We will exploit the connection between entanglement and the length of a Bloch vector

$$\mathbf{a}_i = (\langle s_x \rangle, \langle s_y \rangle, \langle s_z \rangle)$$

$$= a_i \left( \cos(\varphi_i) \sin(\theta_i), \sin(\varphi_i) \sin(\theta_i), \cos(\theta_i) \right)$$

(21)
of a spin. For a classical spin, the length of the Bloch vector is conserved, i.e. $a = 1$. Thus, the total magnetization

$$M = \frac{1}{2N} \sum_i a_i$$

(22)
relaxes only through dephasing of the spins with respect to each other. For spins perfectly aligned in x-direction, this process is inhibited because the state $|X\rangle$ is an eigenstate of the mean-field Hamiltonian. So, for classical spins, the magnetization stays constant. In the quantum realm, this is no longer the case. Both, dTWA and MACE simulations show that the angles $\theta_i$ and $\varphi_i$ are still conserved, but the lengths $a_i \in [-1, 1]$ evolve in time leading to the depolarization dynamics.

For a pure spin-1/2 system $\rho$, the linear entropy $S_{L,i} = 1 - \text{Tr}(\rho_i^2)$ is a measure for the entanglement between the subsystem $\rho_i$ and the rest [41]. In case of the subsystem being an individual spin, the linear entropy can be directly expressed as a function of the length of the Bloch vector $a_i$ via $S_{L,i} = 1 - a_i^2$. A linear entropy of $S_L = 0$ (a length of the Bloch vector $a = 1$) represents a pure quantum state, a linear entropy of $S_L = 1/2$ or a length of the Bloch vector $a = 0$ a completely mixed state. Since the depolarization dynamics are solely given by a variation of the length of the Bloch vector, the linear entropy provides a link between the magnetization and the build-up of entanglement (see also Figure 1).

The blue line in Figure 12 shows the average linear entropy calculated using MACE. Similar to the slow relaxation dynamics of the magnetization, the build-up of entanglement is slow and stretched over multiple decades. Using (22) and the assumption of all
spins being permanently aligned, we can define upper and lower bounds for the average linear entropy $\frac{1}{2}(1 - \frac{1}{N}\sum_i (\alpha_i)^2)$. Given a fixed magnetization, the upper bound is given if all spins have equal length, while the lower bound is determined for all spins either fully polarized or depolarized. For the lower bound, the additional assumptions of all spins pointing in positive direction needs to be fulfilled which holds only for early times (see Figure 12). It should be noted, that a magnetization smaller than 0.5 can only be explained by the build-up of entanglement, assuming that the system is globally in a pure state.

**Summary of parameters**

Table I summarizes the parameters of the individual measurements shown in figure 2 and 3 of the main text.

| fig. | $t_{exc}$ [µs] | $\rho_0^g$ [10¹¹ cm⁻³] | $\rho_0^S$ [10⁹ cm⁻³] | $N_S$ | $R_{bl}$ [µm] | $a$ | $\left(\frac{a}{\rho_0}\right)^{-3}$ | $C_6/(2\pi)/a^6$ | $\beta$ |
|------|----------------|--------------------------|-----------------------|-------|----------------|-----|--------------------------------|-----------------|-------|
| 2, 3 | 1.0            | 1.79(9)                  | 1.2(3)                | 1.2(3)| 5.21          | 5.8(5)| 0.7(2)                       | 1.5(8)          | 0.32(2)|
|      | 0.6            | 1.69(12)                 | 0.43(11)             | 0.4(1)| 4.81          | 8.2(7)| 0.20(5)                      | 0.195(97)       | 0.37(2)|
|      | 1.0            | 1.64(15)                 | 1.2(3)               | 1.1(3)| 5.21          | 5.9(5)| 0.7(2)                       | 1.4(7)          | 0.305(14)|

**TABLE I. Experimental parameters.** $t_{exc}$ denotes the time of laser excitation from the ground to the Rydberg state. $\rho_0^g$ denotes the measured ground state density. $\rho_0^S$ the derived peak spin density. $N_S$, the derived number of total spins. $R_{bl}$ is the blockade radius derived from the excitation time and laser coupling strength. $a$ denotes the Wigner-Seitz radius, $C_6/a^6$ the van der Waals Coefficient, $D_{KL}$ the Kullback-Leibler divergence and $\beta$ is the exponent of the stretched exponential derived from a fit to the relaxation curves.
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