Fast spin squeezing by distance-selective long-range interactions with Rydberg molecule dressing

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We propose a Rydberg molecule dressing scheme to create strong and long-ranged interactions at selective distances. This is achieved through laser coupling ground-state atoms off-resonantly to an attractive molecular curve of two interacting Rydberg atoms. Although dephasing due to Rydberg state decay occurs in all dressing schemes, an advantage of the molecule dressing is that a large ratio of dressed interaction to dephasing rate can be realized at large atomic separations. In an optical lattice or tweezer setting, we show that the strong interaction permits the fast generation of spin squeezing for several tens of dressed atoms. The proposed setting offers a new route to study complex many-body dynamics and to realize quantum information processing with non-convex long-range interactions.

Introduction. The long lifetime and strong interaction emerging between atoms excited to Rydberg states offer an effective way for the implementation of quantum computation protocols [1–10], and many-body quantum simulation [11–20]. One approach to utilize the Rydberg interaction is through the Rydberg dressing [21–32], where ground state atoms are off-resonantly coupled to a Rydberg state. As direct Rydberg excitation is avoided, the dressed interaction - although weakened in strength - comes in principle with the benefit of prolonged coherence times. For Rydberg atoms interacting with a van der Waals potential, the dressing leads to a soft-core interaction (with characteristic soft-core radius $R_c$) between the dressed atoms. This interaction is nearly a constant when the distance $R$ between the atoms is much smaller than $R_c$ and it decays rapidly when $R > R_c$. The strength and $R_c$ of the dressed interaction can be controlled by the Rydberg dressing laser (i.e. detuning, Rabi frequency and the coupled Rydberg state). Such tunability [33, 34] has stimulated the study of exotic topological states [13, 14, 17, 35], density-wave orders [36–38], correlated spin dynamics [39, 40], and quantum sensing [41–43] with Rydberg dressed interactions.

Dissipation [44–48], such as spontaneous decay due to the finite lifetime of Rydberg states [49] and motional dephasing [50–52], is ubiquitous in any Rydberg dressing scheme. In the weak Rydberg dressing scheme (i.e. $|\Delta| \gg \Omega$ with $\Delta$ and $\Omega$ the detuning and Rabi frequency of the dressing laser) [6, 22, 23, 27], the effective decay rate of the dressed state scales with $\sim \Omega^2/\Delta^2$ with $\gamma$ to be the spontaneous decay rate in Rydberg states. As a result, the effective lifetime of the dressed atom is increased by a factor $\Delta^2/\Omega^2$. The strength of the dressing interaction scales as $V_0 \sim \Omega^3\Delta^{-3}$. Its coherence strength is given by the ratio between $V_0$ and the effective decay rate, $C \propto \Omega^2/(\Delta\gamma)$ [27, 53, 54]. These estimations, however, are based on a simple two-level description. There are other detrimental processes such as level crossings at short distances that have to be taken care of in an experimental implementation [27]. To effectively implement quantum simulation and computation with the dressed long-range interactions [55–57], it is worth to developing new Rydberg dressing schemes where such unwanted effects can be suppressed, and which offers a fundamentally better scaling of the coherence strength $C$.

In this work, we propose a Rydberg dressing scheme where ground state atoms are coupled off-resonantly to macroscopic Rydberg molecular dimers which form attractive potentials at micrometer separations [58]. This results in a long-range dressed interaction that is strongly attractive only at selective distances, while saturating to $V_0$ at short distances ($R \ll R_c$). The selective distance locates at the minimal of the molecular potential, whose depth and therefore the dressed interaction can be modulated by external microwave fields. Though laser dressing to Rydberg states generally leads to single- and two-body dephasing, we show that the Rydberg molecule dressing (RMD), however, greatly enhances $C$. As an application, we study spin squeezing of the Rydberg dressed atom in an one dimensional optical lattice. We show that this allows to implement stronger spin squeezing within a much shorter time in comparison to ”traditional” soft-core Rydberg dressing (SRD) [22, 23], provided that the atoms are positioned at certain preselected distances. Our study reveals insights on the coherent and dissipative features in Rydberg gases, which are important for implementing many-body physics and quantum information processing with Rydberg dressed interactions [11–14, 17, 18].

Rydberg molecule dressing. In our setting, each
atom is modeled by two hyperfine ground states $|0\rangle$ and $|1\rangle$, coupled through a Raman process with strength $g$, as shown in Fig. 1(a). A microwave field with the Rabi frequency $\Omega_m$ and the detuning $\Delta_m$ couples Rydberg $nS$ and $n'P$ states (where $n$ and $n'$ are principal quantum numbers) [59]. This leads to a Rydberg superposition state $|\psi\rangle$. The dispersion coefficients of the two Rydberg states are assumed to have opposite signs. In this case, two atoms in the superposition state $|\psi\rangle$ experience an interaction potential $U(R)$ with an attractive tail and a repulsive core, depicted in Fig. 1(b). More details can be found in the supplementary material (SM) [60]. The minimum (located at $R_c$) of $U(R)$ can be a few micrometers, supporting exotic many-body states and macroscopic dimers [58].

In the proposed RMD scheme, the hyperfine state $|1\rangle$ is coupled to state $|\psi\rangle$ where the laser is red-detuned with respect to the potential minimum, as illustrated in Fig. 1(b). When $\Delta \gg \Omega$ and the two-photon detuning $\Delta_2(R) = U(R) + 2\Delta$ is much larger than the effective Rabi frequency $\Omega_2 \equiv \sqrt{2\Omega}^2/2\Delta$ of the collective transition $|11\rangle \leftrightarrow |rr\rangle$ (via the intermediate state $|1r\rangle_s \equiv ((|1r\rangle + |r1\rangle)/\sqrt{2})$, i.e. $|\Delta_2(R_c)| \gg \Omega$, we obtain an effective interaction potential between two atoms in the dressed $|1\rangle$ state,

$$V(R) = \frac{V_0\Delta_2(R)}{\Delta_2^2(R) + \gamma^2} U(R),$$

(1)

with $V_0 = \Omega^4/8\Delta^3$ and $\gamma$ being the spontaneous decay rate in state $|r\rangle$ [60]. As $|\Delta_2(R)| \gg \gamma$, the dressed potential can be further approximated to be $V(R) \approx V_0U(R)/\Delta_2(R)$. The dressed potential is repulsive and attractive at short and large distances [see Fig. 1(c)], respectively. Its depth is largest around the distance $R_c$ determined by $U(R_c)/\Delta_2(R_c)$. Note that the interaction strength at $R_c$ can be several times larger than the saturated value ($\sim V_0$) at short distances. Such profile is different from the soft-core potential [26, 27, 53, 54].

The Rydberg dressing induces not only the dispersive interaction, but also dissipation, whose origin lies in the finite lifetimes in Rydberg states. In our system, the Rydberg dressing laser induces a single-body dephasing (SBD) with the rate $\gamma^{(1)} = \Omega^2 \gamma/4\Delta^2$ and a distance dependent two-body dephasing (TBD) with a rate given by

$$\gamma^{(2)}(R) = \frac{\Omega^4 \gamma}{2\Delta^2(\Delta_2^2(R) + \gamma^2)}.$$ (2)

This two-body dephasing rate reaches the maximal value at $R = R_c$.

Dephasing is ubiquitous in any Rydberg dressing scheme. However, the advantage of the RMD is that the interaction strength $V(R)$ can be enhanced by an order of magnitude with the dephasing only being increased by about a factor of two. To illustrate this, we define the interaction coherence strength $C$

$$C \equiv \frac{V(R)}{2\gamma^{(1)} + \gamma^{(2)}(R)} \approx \frac{\Omega^2}{4\gamma} \frac{\Delta_2(R) U(R)}{\Omega^2 + \Delta_2^2(R)},$$ (3)

where have taken into account the SBD and TBD. The value of $C$ strongly depends on the laser parameters. When $|\Delta_2(R)| \sim |\Delta| \gg \Omega$, one finds the familiar scaling for the SBD $C_s = V_0/2\gamma^{(1)} = \Omega^2/4|\Delta|\gamma$. For $|\Delta| \gg |\Delta_2(R)| \sim \Omega$, the ratio becomes $C = |\Delta_2(R)|/4|\gamma|$, which can be large. For example, we can choose $|\Delta_2(R)| = \Omega$, such that $C_m = \Omega/4\gamma \sim |\Delta|/4\Omega C_s$. As typically $|\Delta| \gg \Omega$, one can gain an order of magnitude increasing in $C$ at $R = R_c$, depicted in Fig. 1(c).

**Effective many-body spin model of dressed Rydberg atoms.** We now consider the many-body dynamics of the atoms trapped in a deep one-dimensional (1D) optical lattice [61, 62]. The dynamics with the dressed ground state manifold is then described by Hamiltonian

$$\mathcal{H}_s = \sum_{k=1}^N [g J_z^{(k)} + \delta(k) J_z^{(k)}] + \sum_{k<l} V(R_{kl}) J_z^{(k)} J_z^{(l)}$$

with the spin-1/2 Pauli operators $J_x^{(k)} = (|1k\rangle\langle0k| + |0k\rangle\langle1k|)/2, J_y^{(k)} = i(|0k\rangle\langle1k| - |1k\rangle\langle0k|)/2,$
The numerical simulations show that the dephasing plays the role of the collective spin operator \( J_z \) as found from our numerical simulation. The dominant contribution results from the SBD, but only become important in the long-time dynamics, seen from the population dynamics, where the master equation (blue solid) by turning off both the SBD and TBD. Other parameters are \( \Omega = 1, (\Delta \gamma \gamma)/\Omega = (5.5, 0.005), R_o = a, g/V_0 = 0.2, n = 50, \) and \( n' = 58 \).

and \( J_z^{(k)} = |1_k\rangle \langle 1_k| - |0_k\rangle \langle 0_k|/2 \). The dressing-induced interaction between atoms located on lattice sites \( k \) and \( l \) at a distance \( R_{kl}/a = |k - l| \) is given by \( V(R_{kl}) \). Taking into account of the single- and two-body dephasing, the dynamics of the many-body two-level system is governed by the master equation,

\[
\dot{\rho} = -i[H_s, \rho] + \sum_{k=1}^{N} L_{\rho|k}\rho + \sum_{k<l} L_{\psi|kl}\rho, \tag{4}
\]

where the Lindblad operator \( L_{\rho|k}\rho = o_{k|\rho} - \frac{1}{2}\{o^\dagger_{\rho|k}, o_{k|\rho}\} \) with \( o_k = \sqrt{\gamma^{(1)}|1_k\rangle \langle 1_k|} \), and \( o_{kl} = \sqrt{\gamma^{(2)}(R_{kl})}|1_k1_l\rangle \langle 1_k1_l| \) being the so-called jump operator corresponding to single- and two-body dephasing, respectively.

We first explore the competition between the dephasing and dressed two-body interaction with an example of two atoms, shown in Fig. 2(a)-(c). During the early stage of dynamical evolution \( 0 < V_0 t < 10 \), both the SBD and TBD hardly affect the coherent dynamics. This is seen from the population dynamics, where the master equation simulation agrees with the coherent dynamics. They only become important in the long-time dynamics, where the many-body coherence is damped. In this region, the dominant contribution results from the SBD, as found from our numerical simulation.

The short-time dynamics is largely coherent even for a moderate number of spins. In Fig. 2, the expectation values of the collective spin operator \( J_z = \sum_k J_z^{(k)} \) and \( J_z = \sum_k J_z^{(k)} \), and the quantum fluctuation \( \langle (\Delta J_z)^2 \rangle \) of the operator \( J_z \) for a chain of \( N = 7 \) atoms are shown. The numerical simulations show that the dephasing plays visible roles when \( V_0 t > 4 \). When \( V_0 t < 4 \), the high coherence strength guarantees that the dispersive interaction dominates in the short-time dynamics.

**Fast spin squeezing.** We will show that the large \( C \) in the RMD allows to achieve rapid spin squeezing in the 1D lattice [63]. The degree of spin squeezing can be quantified through the squeezing parameter (SP) [64],

\[
\xi^2 = \frac{N(\Delta J_{\perp}^2)}{\langle |J|^2 \rangle}, \tag{5}
\]

where \( J_{\perp} \) is the spin component perpendicular to the direction of the mean total spin \( \langle J \rangle \) with \( \langle J \rangle = \sum_k J^{(k)} \).

The parameter \( \xi^2 \) is then defined by choosing the direction \( \vec{n}_1 \) at which \( \Delta J_{\perp} \) is minimized. For the state with \( \xi^2 < 1 \), its phase sensitivity is improved over the standard quantum limit \( \sim 1/\sqrt{N} \) given by the coherent spin state [65].

To achieve spin squeezing, initially the atoms are prepared in the state \( |0\rangle \) such that the mean spin direction is along the \( z \)-direction, and thus the perpendicular spin component is simply \( J_{\perp} = \cos(\theta) J_x + \sin(\theta) J_y \) with \( \theta \) being the angle with respect to the \( x \) axis, which immediately implies \( (\Delta J_{\perp}^2)_{\perp} = \cos^2(\theta) \langle J_x^2 \rangle + \sin^2(\theta) \langle J_y^2 \rangle + \sin(2\theta) \langle J_x J_y \rangle + J_x J_y / 2 \). We then implement the spin-echo type squeezing protocol [42], which consists of a \( \pi/2 \) pulse of length \( \tau_2/2 = \pi/(2g) \) to rotate the spin along the \( x \) axis, RMD of duration \( \tau/2 \), a \( \pi \) pulse to rotate the spin, RMD of duration \( \tau/2 \), and then a \( \pi/2 \) pulse. During the spin rotation, the Rydberg dressing laser is turned off. At the end of the sequence, the mean spin is along the \( z \)-direction again with the noise uncertainty for \( J_{\perp} \), along one of the perpendicular direction being squeezed.

The SP resulting from the above protocol can be analytical calculated in the absence of dissipation [42]. In the presence of dissipation, it is not possible to carry out analytical calculations for the dressed spin model. For small systems with a few atoms we can study the dressed-dissipative dynamics by numerically solving the master equation and evaluate the SP. Approximations have to be taken when there are tens and even hundreds of lattice sites due to the large Hilbert space. Since the \( \pi/2 \) and \( \pi \) rotational pulses are much shorter than the typical time scale of the SBD and TBD, dissipation is therefore not important during the dynamical evolution. This allows us to simulate the dynamics approximately with a non-Hermitian Hamiltonian \( \mathcal{H}_e = \mathcal{H}_s - i(\gamma^{(1)}/2) \sum_{k=1}^{N} |1_k\rangle \langle 1_k| - (i/2) \sum_{j<k} \gamma^{(2)}(R_{jk})|1_k 1_j\rangle \langle 1_j 1_k| \), where the single- and two-body jump processes have been neglected. Furthermore, we apply the mean-field approximation to the decoherence part induced by the TBD, and finally arrive at...
the non-Hermitian Hamiltonian
\[ H_c \approx \frac{1}{2} \sum_{j=1}^{N} \left( \sum_{k \neq j} V(R_{jk}) - i \Gamma_z \right) J_z^{(j)} + \sum_{j < k} V(R_{jk}) J_z^{(j)} J_z^{(k)} - i \frac{\bar{\Gamma}}{2}, \tag{6} \]

where the mean-field decay rate \( \bar{\Gamma} = N \left[ \gamma^{(1)} + \Gamma_0 \left( \frac{1}{4} - \langle J_z \rangle^2/N^2 \right) \right] \) and \( \Gamma_z = \gamma^{(1)} + \Gamma_0 \left( \frac{1}{2} + \langle J_z \rangle/N \right) \) with \( \Gamma_0 = \sum_{j=1}^{N-1} \gamma^{(2)}(R_{jk}). \) We then evaluate the time-dependent many-body state \( \langle \psi(t) \rangle \approx e^{-i H_c(t)} |\psi(0)\rangle \) where the initial state \( |\psi(0)\rangle \) corresponds to all atoms in state \( |0\rangle. \)

Eq. (6) enables us to find an analytical solution in the presence of the dephasing and for large systems [see SM [60]]. To verify the accuracy of the analytical solution against the exact result, we first examine a situation with \( N = 10 \) atoms with \( (\Delta, \gamma)/\Omega = (10, 0.005) \) (corresponding to \( \gamma^{(1)}/\gamma \approx 8.3 \times 10^{-3} \)), depicted in Fig. 3. For \( R_c = a \), both the effective master equation [66, 67] and the non-Hermitian Hamiltonian give the similar optimal SP (i.e. the minimum) \( \xi^2_{\text{min}} \sim 0.6 \) around the dressing time \( V_0 \tau_{\text{min}} \approx 0.17 \), which is much shorter than that by the traditional SRD scheme [42], and see Fig. 4(a). The dissipative SP is slightly weaker than the coherent counterpart \( \xi^2 \sim 0.58 \) without including the decoherence. For \( R_c = 3a \), because the repulsive interactions among the nearest and the second-nearest neighboring sites counteract the effect of the strong attractive interac-
tions, the optimal SP reduces to \( \xi^2_{\text{min}} \sim 0.83 \). On the hand, in comparison with the individually spontaneous decay, the TBD does not cause significant effects on the rapid spin squeezing at the time scale \( V_0 \tau_{\text{min}} \). We find that the dissipative dynamics is well captured by the conditional Hamiltonian \( H_c \) in general, which motivates us to investigate larger systems with \( H_c \) (see SM [60]).

Based on Eq. (6), it is not difficult to find that the time-dependent squeeze parameter \( \xi^2(\tau) \) exhibits an exponential decay as the dressing time \( \tau \) increases, namely,
\[ \xi^2(\tau) \propto e^{-\bar{\Gamma} \tau}, \]
with \( \bar{\Gamma} \) depending on the effective single-and two-body dephasing rates. We show in Fig. 4 the optimally dissipative SP \( \xi^2_{\text{min}} \) and the corresponding dressing time \( V_0 \tau_{\text{min}} \) versus the number of the lattice sites with different lattice constant \( R_c = ka \) \( (k = 1, 2, 3) \). For comparison, we also show the SP influenced only by the SRD, where the TBD is negligible [42]. In both cases, the detrimental effects of the dephasing are to diminish the squeezing when the number of atoms increases. In particular, the spin squeezing is more robust in case of RMD, where the SP is smaller than 1 for \( N \) up to 200 [see the first and third row in Fig. 4]. In contrast the SP reaches 1 typically at smaller \( N \) with the SRD. On the other hand, for the RMD scheme with \( R_c = a \), where the dressed atoms next to each other get the strongest interaction coherence strength \( C \), the optimal SP exists for \( N > 150 \) and the dressing times are all within \( V_0 \tau_{\text{min}} < 0.17 \); the SRD scheme however allows for \( \xi^2_{\text{min}} < 1 \) when the system contains only a few tens of atoms and requires a much longer dressing time. As the lattice constant decreases from \( a = R_c/2 \) to \( R_c/3 \), the SP for the RMD scheme decreases. This reduction results from the fact that the phase of the many-body state \( |\psi(t)\rangle \) due to
repulsive \([V(R) > 0]\) and attractive \([V(R) < 0]\) interactions cancels. In contrast, the SRD scheme becomes more robust to the dephasing for \(a = R_c/3\) due to the constructive phase accumulation by all repulsive interactions (i.e. the interaction potential is positive for all realized interatomic distances). In general, a compromise could be found in between getting a large interaction coherence strength and keeping all negative (or positive) phase terms. In this regard, the SP under the RMD can be better than that by the SRD for \(N > 40\) and \(R_c = 2a\), but the SRD scheme is able to achieve strong SP, i.e., \(\xi_{\text{min}}^2 < 0.5\) for \(R_c > 3a\) and \(N < 30\).

**Conclusion.**—We have proposed a Rydberg molecule dressing scheme, where the dressing laser is tuned off-resonantly to be the attractive molecular potential of two Rydberg atoms, which can be created by coupling two different Rydberg states with MW fields. The Rydberg molecule dressing leads to distance-selective attractive interaction potential of the Rydberg dressed ground-state atoms. We have shown that enhanced Rydberg dressed interactions can be achieved, while the respective dissipation is still weak. This allows us to generate entangling phases for fast spin squeezing. Our work is relevant to the study of quantum computation and simulation with finite systems of Rydberg atoms trapped in optical arrays [68–70] and trapped Rydberg ions [71]. In the future, it is worth to explore squeezing in higher dimensional settings and investigate whether the distance selective two-body interaction also here lead to enhanced squeezing. The controllable interaction and dephasing furthermore opens opportunities to quantum simulate interesting many-body dynamics, complementary to other studies that aim at the creation of exotic phases and many-body states [72–75]. We note that a recent experiment has demonstrated the distance selective interaction in an atom array [76], where the ground state atom is dressed to Rydberg macrodimers [77]. Our scheme offers the flexibility to engineer the dressed interaction through tuning the MW field [58].

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Supplementary material for: “Fast spin squeezing by distance-selective long-range interactions with Rydberg molecule dressing”

This supplementary material contains additional details on the analysis in the main text.

**MICROWAVE MODULATED RYDBERG INTERACTION ENERGIES AND RYDBERG MOLECULAR DRESSING**

We consider two interacting Rydberg atoms separated by $R$, where two Rydberg states $|np\rangle$ and $|n's\rangle$ are continuously driven by a microwave (MW) field with Rabi frequency $\Omega_{mw}$ and detuning $\Delta_{mw}$. The Rydberg interaction between the states $|np\rangle$ and $|n's\rangle$ is of the dipole nature and scales as $R^{-3}$, while the van der Waals interaction scaling as $R^{-6}$ takes over if the atoms simultaneously populate the same Rydberg level $|n's\rangle$ (or $|np\rangle$). The Hamiltonian for the coupled Rydberg pair states $\{|n's,n's'\},\{|n's,np\} + |np,n's\rangle)/\sqrt{2}, |np,np\rangle}$ reads

$$H_{sp}(R) = \begin{bmatrix} -\frac{C_{(s,s)}^{I(s,s)}}{\hbar \omega} & \frac{\Omega_{mw}}{\sqrt{2}} & 0 \\ \frac{\Omega_{mw}}{\sqrt{2}} & \Delta_{mw} + \frac{C_{(s,p)}^{I(s,p)}}{\hbar \omega} & \frac{\Omega_{mw}}{\sqrt{2}} \\ 0 & 2\Delta_{mw} - \frac{C_{(p,p)}^{I(p,p)}}{\hbar \omega} & 0 \end{bmatrix}. \quad (S1)$$

The eigenenergies $E_{n}^{(2)}(R)$ $(n = 1,2,3)$ of the Hamiltonian $(S1)$ give the modified level configuration (e.g. the eigenstates $|\phi_{n}^{(2)}\rangle$) for the Rydberg pair states, which strongly depend on the coupling strength and detuning between the MW field and the Rydberg states. As a specific example, we show, in Fig. S1(a), the distance-dependent interactions between the Rydberg states $|58p\rangle$ and $|50s\rangle$ for Rb atoms. The Rydberg molecular potential appears at the upper (black solid) branch corresponding to the pair state $|50s\rangle + |50s\rangle$ at large distances. It shows an attractive character when the interatomic separation $R$ is larger than 2 $\mu$m and is repulsive at short distances.

The ground state atoms are coupled from the ground state $|5s\rangle$ to the MW coupled Rydberg states $|E_{1}^{(1)}\rangle = \sin(\phi/2)|n's\rangle + \cos(\phi/2)|np\rangle$ ($|E_{1}^{(1)}\rangle = -\sin(\phi/2)|np\rangle + \cos(\phi/2)|n's\rangle$) with the laser detunings $\Delta_{+}(\Delta_{-})$ and coupling strengths $\Omega_{+} = \cos(\phi/2)\hat{d}_{0}, \hat{E}_{0}/\hbar$ ($\Omega_{-} = \sin(\phi/2)\hat{d}_{0}, \hat{E}_{0}/\hbar$), where $\phi = tan^{-1}(2\Omega_{mw}/\Delta_{mw})$, $\hat{d}_{0}$ is the dipole moment of the transition $|5s\rangle \leftrightarrow |np\rangle$, and $\hat{E}_{0}$ is the electric field amplitude. We are interested in the RMD based on two-photon dispersive interactions, that is, the doubly-excitation states are far-off resonant from the two-photon transition mediated by $|E_{1}^{(1)}\rangle$. At large distances, the effective Rabi frequencies for the transition channel $|5s\rangle|5s\rangle \rightarrow |5s\rangle|E_{1}^{(1)}\rangle \rightarrow |\phi_{1}^{(2)}\rangle$ ($\sim |E_{1}^{(1)}\rangle|E_{1}^{(1)}\rangle$) is approximately given by $\Omega_{2} \approx \Omega_{2}^{2}/\Delta_{+}$. While at short distances, $\Omega_{2}(R)$ becomes distance-dependent. The parameter regime for RMD is thus given by $|\Delta_{+}| \gg \Omega_{+}$ and $|2\Delta_{+} + E_{1}^{(1)}(\infty) - E_{1}^{(2)}(R)| \gg \Omega_{2}, \gamma$, from which we can then find the dressed interaction potential via the perturbation theory (see the following section). The perturbation result of the dressed interaction potential agrees with the full numerical calculation, shown in Fig. S1(b).

**DERIVATION OF THE EFFECTIVE MASTER EQUATION**

We consider two hyperfine ground states $|0\rangle$ and $|1\rangle$ where state $|1\rangle$ is weakly coupled to the Rydberg state $|r\rangle = |+\rangle$ by the dressing laser (Rabi frequency $\Omega = \Omega_{+}$ and detuning $\Delta = \Delta_{+}$). The Hamiltonian of the atoms is given by $H_{a} = H_{a} + H_{t}$, where the atomic Hamiltonian for the spin states $|0\rangle, |1\rangle$ and Rydberg state $|r\rangle$ is

$$H_{a} = \sum_{j} \left[ \Delta_{a} \sigma^{(j)}_{rr} + \Delta_{0} \sigma^{(j)}_{00} + \left( \frac{g}{2} \sigma^{(j)}_{01} + \frac{\Omega}{2} \sigma^{(j)}_{1r} + H.c. \right) \right], \quad (S2)$$

where $\Delta_{0}$ and $g$ is the detuning and Rabi frequency between the two hyperfine states (e.g. coupled through a MW field or a Raman process). The two-body interaction in the Rydberg state is

$$H_{i} = \frac{1}{2} \sum_{jk} U(R_{jk}) \sigma^{(j)}_{rr} \sigma^{(k)}_{rr}, \quad (S3)$$
where \( U(R) = E_2^{(2)} \) is the molecular interaction potential of the MW coupled Rydberg state. Taking into account of the spontaneous decay in the Rydberg state, the dynamics is governed by a master equation

\[
\dot{\hat{\rho}} = -[H_0, \hat{\rho}] - \gamma \sum_j \left( \sigma_{1r}^{(j)} \rho \sigma_{1r}^{(j)} - \frac{1}{2} \{ \sigma_{1r}^{(j)} \sigma_{1r}^{(j)} \rho \} \right).
\]  

(S4)

We first derive the effective master equation of two atoms. This is done in a regime where the detuning of the dressing laser is large, i.e. \( \Delta > \Omega \gg \gamma \approx \Delta_0 \). We will then eliminate dynamics in the Rydberg state to obtain the effective dynamics by separating different time scales. As the dynamics due to the dressing laser is fast, this allows us to split the Hilbert space into two parts, \( P \) and \( Q \) where \( P \)- and \( Q \)-subspace define the slowly and rapidly varying quantities with \( P + Q = i\mathbb{I} \). The effective Hamiltonian for the slow dynamics can be obtained through

\[
H_{\text{eff}} = PHP - PHQ \frac{1}{QHQ} QHP.
\]  

(S5)

In the two atom basis \( \{ |00\rangle, |01\rangle, |10\rangle, |11\rangle, |rr\rangle \} \), the matrix form of the Hamiltonian is expressed as

\[
H_{\text{eff}} = \begin{pmatrix}
2\Delta_0 & 0 & 0 & 0 & 0 \\
0 & \Delta_0 - \frac{\Omega^2}{2\Delta} & 0 & 0 & 0 \\
0 & 0 & \Delta_0 - \frac{\Omega^2}{2\Delta} & 0 & 0 \\
0 & 0 & 0 & \frac{\Omega^2}{2\Delta} & -\frac{\Omega^2}{2\Delta} \\
0 & 0 & 0 & \frac{\Omega^2}{2\Delta} & 2\Delta - \frac{\Omega^2}{2\Delta}
\end{pmatrix}
\]

To study the dissipative process, we assume decay of Rydberg states is fast than the coherent couplings. To consider different time scales, the master equation \( \dot{\hat{\rho}} = (\mathcal{L}_0 + \mathcal{L}_1)\hat{\rho} \) is split into the fast (denoted by \( \mathcal{L}_0\hat{\rho} \)) and slow (denoted by \( \mathcal{L}_1\hat{\rho} \)) parts, where

\[
\mathcal{L}_0\hat{\rho} = \sum_{j=1,2} \frac{1}{\gamma} \left( \sigma_{1r}^{(j)} \hat{\rho} \sigma_{1r}^{(j)} - \frac{1}{2} \{ \hat{\rho}, \sigma_{1r}^{(j)} \sigma_{1r}^{(j)} \} \right),
\]

\[
\mathcal{L}_1\hat{\rho} = -i[\mathcal{H}_{\text{eff}}, \hat{\rho}].
\]  

(S6)

We will trace the fast dynamics and derive an effective master equation for the slow dynamics via the second order perturbation calculation.

Here we define a projection operator \( \mathcal{P}_0 = \lim_{t \to \infty} e^{t\mathcal{L}_0} \), which projects the density matrix to the subspace corresponding to the relatively slow dynamics, i.e. \( \hat{\rho}_0 = \mathcal{P}_0\hat{\rho} \). The first order correction to the slowly varying dynamics vanishes \( \mathcal{P}_0\mathcal{L}_1\mathcal{P}_0\hat{\rho} = 0 \). We then calculate the second order correction \( -\mathcal{P}_0\mathcal{L}_1(1-\mathcal{P}_0)\mathcal{L}_1\mathcal{P}_0\hat{\rho} \). A tedious but straightforward calculation yields an effective master equation depending on the two-atom dephasing,

\[
\dot{\hat{\rho}}_c \approx -i[\tilde{V}_{12}, \hat{\rho}_c] + \gamma_{12} \left( \sigma_{11}^2 \sigma_{11}^2 \hat{\rho}_c \sigma_{11}^2 \sigma_{11}^1 - \frac{1}{2} \{ \sigma_{11}^2 \sigma_{11}^1 \hat{\rho}_c \} \right).
\]  

(S7)
where \( \gamma_{12} = \Omega^4\gamma/2\Delta^2[(U(R_{12}) + 2\Delta)^2 + \gamma^2] \) and the dressed interaction \( V_{12} = \Omega^4[U(R_{12}) + 2\Delta]/2\Delta^2[(U(R_{12}) + 2\Delta)^2 + \gamma^2] \) without including the correction induced by the single-photon Stark shifts.

To illustrate the accuracy of the perturbative calculation, we consider the system dynamics under the Rydberg molecule dressing with \((U(R_{12}), \Delta)/\Omega = (21, 10)\) (left panel) and \((U(R_{12}), \Delta)/\Omega = (40, 10)\) (right panel), respectively, shown in Fig. S2. The former leads to the dressed interaction \( V_{12}/\Omega \approx 2.5 \times 10^{-3} \) and the two-body decay rate \( \gamma_{12} \sim \Omega^2\gamma/2\Delta^2 = 5 \times 10^{-3}\gamma \), which is comparable to the effective single-photon decay \( \gamma_1 \sim \Omega^2\gamma/4\Delta^2 \); while for the latter, the dressed energy and the effective decay rate are \( V_{12}/\Omega \approx 2.5 \times 10^{-4} \) and \( \gamma_{12} \sim 10^{-3}\gamma_1 \), respectively. The dissipative dynamics of the two-atom system is then determined by the ratios with respect to the driving strength: \( g/V_{12} \) and \( g/(2\gamma_1 + \gamma_{12}) \). For the limit \( V_{12}/g \gg 1 \), the system can only transit between the states \(|00\rangle\) and \(|10\rangle\) (|01\rangle), corresponding to the two-photon blockade, and the two-body decay accelerates the convergence to the steady state. In contrast, for \( g/V_{12} \gg 1 \), the two-body decay effect can be largely neglected.

**SPIN SQUEEZING OF THE RYDBERG DRESSED ATOMS**

Consider the effective decay described by the non-Hermitian (conditional) Hamiltonian

\[
H_c = \sum_{i=1}^{N} \left[ \frac{1}{2} \sum_{j \neq i} \pi_{ij} - i \frac{\gamma_j}{2} \right] J_z^{(i)} + \sum_{i<j} \pi_{ij} J_z^{(i)} J_z^{(j)} - i \frac{\Gamma G}{2}, \tag{S8}
\]

\[
H_m = \sum_{i=1}^{N} \left( -\frac{\Delta m}{2} J_z^{(i)} + \Omega_m J_x^{(i)} \right) + \text{H.c.}, \tag{S9}
\]

where

\[
\pi_{ij} = V(R_{ij}) - i \frac{\gamma^{(2)}_{ij}}{2},
\]
where

\[
\Gamma_G = \frac{N\gamma^{(1)}}{2} + \frac{1}{2} \sum_{i<j} \gamma^{(2)}_{ij},
\]

with

\[
\gamma^{(2)}_{ij} = \frac{\Omega^4}{2\Delta^2[(U(R_{ij}) + 2\Delta)^2 + \gamma^2]}
\]

being the distance-dependent two-atom collective decay rate.

Under the mean-field approximation, the collective decay can be rewritten in the following form,

\[
\sum_{j<k} \gamma^{(2)}_{jk} \hat{\sigma}^{(j)}_n \hat{\sigma}^{(k)}_n = \frac{1}{2} \sum_j \gamma^{(2)}_n \left( \langle \hat{\sigma}^{(j)}_n \rangle + \langle \hat{\sigma}^{(j)}_n \hat{\sigma}^{(k)}_n \rangle - \langle \hat{\sigma}^{(j)}_n \rangle \langle \hat{\sigma}^{(k)}_n \rangle \right) + \frac{1}{2} \sum_k \langle \hat{\sigma}^{(j)}_n \rangle \left( \sum_{k\neq j} \gamma^{(2)}_{jk} \langle \hat{\sigma}^{(k)}_n \rangle \right) - \frac{1}{2} \sum_j \langle \hat{\sigma}^{(j)}_n \rangle \left( \sum_{k\neq j} \gamma^{(2)}_n \langle \hat{\sigma}^{(k)}_n \rangle \right)
\]

\[
= \Gamma(0) \langle \hat{\sigma}_n \rangle \sum_j \hat{\sigma}^{(j)}_n - \frac{N}{2} \hat{\Gamma}(0) \langle \hat{\sigma}_n \rangle^2,
\]

(S10)

where \( \hat{\Gamma}(q) \) is the Fourier transform of the dissipation-rate matrix \( \gamma^{(2)}_{jk} = \gamma^{(2)}(|R_j - R_k|) \):

\[
\hat{\Gamma}(q) = \sum_{R} \gamma^{(2)}(R) e^{-i q R}
\]

(S11)

and therefore \( \hat{\Gamma}(0) = \sum_{k-j=1}^{N-1} \gamma^{(2)}_{jk} \), which is denoted as \( \Gamma_0 \) later for conciseness. For nearest neighbor interactions only, one has \( \Gamma_0 = Z\gamma^{(2)}_{12} \), where \( Z \) is the lattice coordination number, i.e. the number of nearest neighbors of any given site. For the one dimensional case, we thus have

\[
H_c = \sum_{i=1}^{N} \left[ -i \frac{\gamma^{(1)}}{2} + \frac{1}{2} \sum_{j \neq i} \left( V_{ij} - i \frac{\gamma^{(2)}_{ij}}{2} \right) \right] J_z^{(i)} + \sum_{i<j} \left( V_{ij} - i \frac{\gamma^{(2)}_{ij}}{2} \right) J_z^{(i)} J_z^{(j)} - i \frac{\Gamma_G}{2},
\]

\[
= \sum_{i=1}^{N} \left( \frac{1}{2} \sum_{j \neq i} V_{ij} \right) J_z^{(i)} + \sum_{i<j} V_{ij} J_z^{(i)} J_z^{(j)} - i \left[ \frac{\gamma^{(1)}}{2} + \frac{\Gamma_0}{4} \right] \sum_{i=1}^{N} J_z^{(i)}
\]

\[
- \frac{i}{2} \left[ \Gamma_0 \langle J_z^{(i)} \rangle \sum_{j} J_z^{(j)} - \frac{N}{2} \Gamma_0 \langle J_z^{(i)} \rangle^2 \right] - i \frac{\Gamma_G}{2}
\]

\[
= \sum_{j=1}^{N} \left( \frac{1}{2} \sum_{k \neq j} V_{jk} \right) J_z^{(j)} + \sum_{j<k} V_{jk} J_z^{(j)} J_z^{(k)} - i N \left[ \frac{\gamma^{(1)}}{4} + \frac{\Gamma_0}{4} \left( \frac{1}{4} - \langle J_z \rangle^2 \right) \right]
\]

\[
- \frac{i}{2} \left[ \gamma^{(1)} + \Gamma_0 \left( \frac{1}{2} + \langle J_z \rangle \right) \right] \sum_{j=1}^{N} J_z^{(j)},
\]

\[
= \sum_{j=1}^{N} \left( \sum_{k \neq j} V_{jk} - i \Gamma_z \right) J_z^{(j)} + \sum_{j<k} V_{jk} J_z^{(j)} J_z^{(k)} - i \frac{\Gamma}{2},
\]

(S12)

where

\[
\bar{\Gamma} = \frac{N}{2} \left[ \gamma^{(1)} + \Gamma_0 \left( \frac{1}{4} - \langle J_z \rangle^2 \right) \right],
\]
\[ \Gamma_z = \gamma^{(1)} + \Gamma_0 \left( \frac{1}{2} + \langle J_z \rangle \right), \]

and \( \langle J_z \rangle \) is the mean value of the \( z \)-component of the angular momentum for the individual atom. For numeric, we evaluate \( \langle J_z \rangle \) by the weighted mean for all spins and the time average for the implementation period \( \tau \), namely,

\[ \overline{\langle J_z(\tau) \rangle} = \frac{1}{N \tau} \int_0^\tau d\tau \langle J_z(\tau) \rangle, \]

which will be analytically calculated in the Heisenberg picture and should be effective in the mean-field regime.

We measure the degree of spin squeezing by the parameter proposed by Wineland et al. \[S64\],

\[ \xi^2 = \frac{N (\Delta J_{\tilde{r}_z})^2}{\langle \tilde{J} \rangle^2}, \] (S13)

where

\[ \langle \tilde{J} \rangle = \left( \sum_{k=1}^{N} J^{(k)} \right) = \sqrt{\langle J_x \rangle^2 + \langle J_y \rangle^2 + \langle J_z \rangle^2} \tilde{\varepsilon}_z \] (S14)

is the mean collective angular momentum of all spins with its direction \( \hat{\varepsilon}_z \) being predefined along the \( z \)-axis, corresponding to the initial unitary population of the ground state for all atoms, and

\[ J_{\tilde{r}_z}(\theta) = \cos(\theta) J_x + \sin(\theta) J_y \] (S15)

is the perpendicular spin component with \( \theta \) being the angle with respect to the \( x \) axis. Thus, the fluctuations in \( J_{\tilde{r}_z}(\theta) \) can be calculated by

\[ (\Delta J_{\tilde{r}_\perp})^2 = \langle J_{\tilde{r}_\perp}^2(\theta) \rangle - \langle J_{\tilde{r}_\perp}(\theta) \rangle^2 \]

\[ = \left( \cos^2(\theta) \langle J_x^2 \rangle + \sin^2(\theta) \langle J_y^2 \rangle + \sin(\theta) \cos(\theta) \langle J_x J_y + J_y J_x \rangle \right) \]

\[ - \left( \cos^2(\theta) \langle J_x \rangle^2 + \sin^2(\theta) \langle J_y \rangle^2 + 2\sin(\theta) \cos(\theta) \langle J_x \rangle \langle J_y \rangle \right), \]

\[ = \cos^2(\theta) (\Delta J_x)^2 + \sin^2(\theta) (\Delta J_y)^2 \]

\[ + \sin(\theta) \cos(\theta) \left( \langle J_x J_y + J_y J_x \rangle - 2 \langle J_x \rangle \langle J_y \rangle \right). \] (S16)

Without considering the effect of the atomic spontaneous emission (i.e., \( \gamma^{(1)} = 0 \) and \( \gamma^{(2)}_{ij} = 0 \)), and based on the spin-echo protocol, one can find mean values of the spin components analytically \[S42\]:

\[ \langle J_x \rangle = \langle J_y \rangle = 0, \quad \langle J_z \rangle = \left( \sum_{i=1}^{N} J_i^{(i)} \right) = -\frac{1}{2} \sum_{i} N \prod_{j \neq i} \cos(\varphi_{ij}), \] (S17)

\[ \langle J_x^2 \rangle = \left( \sum_{k=1}^{N} J_x^{(k)} \right) \cdot \left( \sum_{l=1}^{N} J_x^{(l)} \right) = \frac{N}{4} + 2 \sum_{k<l}^{N} \langle J_x^{(k)} J_x^{(l)} \rangle \]

\[ = \frac{N}{4} + \frac{1}{4} \sum_{i<j}^{N} \left[ \prod_{k \neq i,j}^{N} \cos(\varphi_{i,j,k}) - \prod_{k \neq i,j}^{N} \cos(\varphi_{i,j,k}^+) \right], \] (S18)

\[ \langle J_y^2 \rangle = \frac{N}{4} + 2 \sum_{i<j}^{N} \langle J_y^{(i)} J_y^{(j)} \rangle = \frac{N}{4}, \] (S19)

and

\[ \langle J_x J_y + J_y J_x \rangle = -\sum_{i<j}^{N} \sin(\varphi_{ij}) \prod_{k \neq i,j}^{N} \cos(\varphi_{ik}), \] (S20)
where $\phi_{ij} = V_{ij}\tau/2$ and $\phi_{ijk}^\pm = \phi_{ik} \pm \phi_{jk}$.

When we take into account the non-Hermitian Hamiltonian (S12), where the decoherence is estimated by the mean-field single-body decay, and redo the spin-echo protocol by using the following transformations

\begin{align*}
e^{i\alpha J_x^{(k)} J_y^{(k)} e^{-i\alpha J_x^{(k)}}} &= \cos(\text{Re } \alpha) J_x^{(k)} - \sin(\text{Re } \alpha) J_y^{(k)}, \quad (S21) \\
e^{i\alpha J_y^{(k)} J_y^{(k)} e^{-i\alpha J_y^{(k)}}} &= \cos(\text{Re } \alpha) J_y^{(k)} + \sin(\text{Re } \alpha) J_x^{(k)}, \quad (S22) \\
e^{i\alpha J_z^{(k)} J_z^{(k)} e^{-i\alpha J_z^{(k)}}} &= e^{2i\alpha J_z^{(k)} J_x^{(k)}}, \quad (S23)
\end{align*}

the mean values and the quantum fluctuations of the spin angular momentum operators are alternatively given by

\begin{align*}
\langle J_m \rangle &\rightarrow e^{-\bar{\Gamma}\tau} \langle J_m \rangle, \quad \langle J_m J_n \rangle \rightarrow e^{-\bar{\Gamma}\tau} \langle J_m J_n \rangle, \quad (S24)
\end{align*}

and

\begin{align*}
\langle \Delta J_{\perp} \rangle^2 = &e^{-\bar{\Gamma}\tau} \left[ \cos^2 (\theta) \langle J_x^2 \rangle + \sin^2 (\theta) \langle J_y^2 \rangle \right] \\
&+ \frac{1}{2} \sin (2\theta) \langle J_x J_y + J_y J_x \rangle, \quad (S25)
\end{align*}

where $m, n = x, y, \text{or } z$, $\tau$ is the total optical dressing time, and we have assumed the rotation pulses is fast enough, such that the dissipation is negligible during the operation. Moreover, the dissipative effects in the $z$-component of the individual spin angular momentum $[J_z^{(k)}(\tau) \sim e^{-\bar{\Gamma}\tau}]$ during the two halves of optical dressing time counteract each other due to spin-flip induced by the rotation $\pi$ pulse. Using the above results, we then calculate the squeezing parameter for different number of sites and interactions (RMD and SRD).