

Simulation of XXZ Spin Models using Sideband Transitions in Trapped Bosonic Gases: Supplemental Materials

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S1. EXPERIMENTAL REALIZATION

To initiate our experiments, ultracold ^{87}Rb atoms are captured in a magneto-optical trap loaded from a background gas. The captured atoms are subsequently transported to a vacuum region with reduced pressure, where they are cooled by forced evaporative cooling. Initial cooling is performed in a hybrid trap that combines a magnetic quadrupole trap with an optical dipole potential [S1]. The final temperatures are achieved by evaporation in a pure optical dipole trap. This trap is formed by two laser beams at a wavelength of 1064 nm, which intersect at an angle of 18° as shown in Fig. S1(a). The two beams have waists of 60 and $75\ \mu\text{m}$, which provide a nearly harmonic trapping potential with trapping frequencies of 143 Hz, 21.5 Hz and 171 Hz at the chosen laser power.

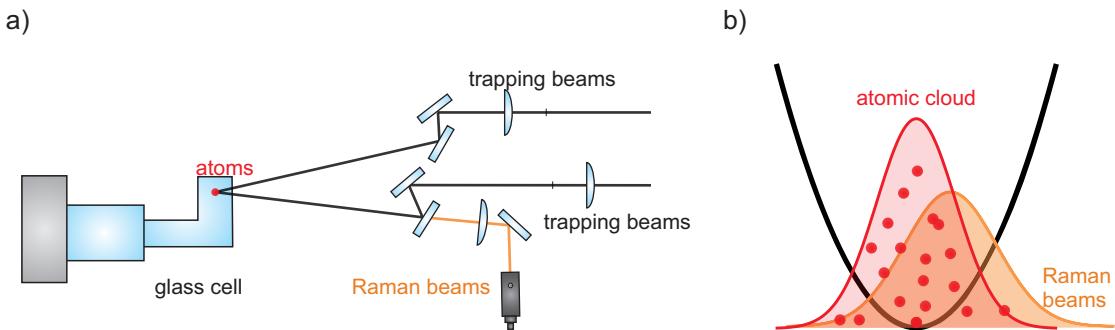


FIG. S1. (a) Experimental implementation. The optical dipole potential is formed by two laser beams (black) which intersect with an angle of 18° in the experimental chamber. The Raman laser beams (orange) are directed onto the atomic cloud along one of these beams. (b) To address the motional sidebands an asymmetric coupling field is required, which is realized by introducing a small offset between the Raman laser focus and the atomic cloud.

The procedure outlined above results in an ensemble of 4×10^5 atoms in the $|F=2, m_F=2\rangle$ state at a temperature of $375(25)$ nK. Subsequently, the atoms are transferred to the $|\uparrow\rangle = |F=2, m_F=0\rangle$ state by a radio-frequency rapid adiabatic passage. In a final step, the atomic ensemble is initialized with a variable mean density n between 0.46 and $4.8 \times 10^{12}\text{cm}^{-3}$ in the $|\downarrow\rangle = |F=1, m_F=0\rangle$ state by a microwave Rabi pulse of variable duration. The remaining atoms in the $|\uparrow\rangle$ state are removed with a resonant light pulse on an optical transition. Importantly, this method allows for the preparation of a variable density at constant temperature. The density of these ensembles is calibrated by performing microwave-based Ramsey interferometry on the clock transition (Fig. S2(b)) and by recording the density-dependent frequency shift of the carrier transition ($-0.48\text{Hz}/10^{12}\text{cm}^{-3}$ [S2]). Here we provide a list that connects the mean atom density n and the corresponding atom number N we used in the DPT experiment:

Mean atom density n (cm^{-3})	0.46×10^{12}	0.98×10^{12}	2.0×10^{12}	3.2×10^{12}	4.8×10^{12}
Atom number N	0.33×10^5	0.69×10^5	1.4×10^5	2.2×10^5	3.4×10^5

The inhomogeneous coupling field between the two clock states $|\downarrow\rangle$ and $|\uparrow\rangle$ is realized by using two copropagating Raman beams which are derived from two phase-locked diode lasers. The Raman beams are focused onto the atomic cloud with a beam waist of $39\ \mu\text{m}$. In principle, these Raman beams can lead to a differential shift of the clock states and thus fluctuations in the Raman beam power may lead to a significant broadening of the spectroscopic signal. To avoid this effect, a specific relative intensity of the Raman beams can be chosen, which reduces the light shift [S3]. Figure S2(a)) shows the experimental determination of the optimal relative intensity based on the comparison of spectroscopy on the clock transition with the Raman system and with microwave radiation. In the experiments a relative intensity $I_1/I_2 = 1.98$ was chosen to avoid differential shifts.

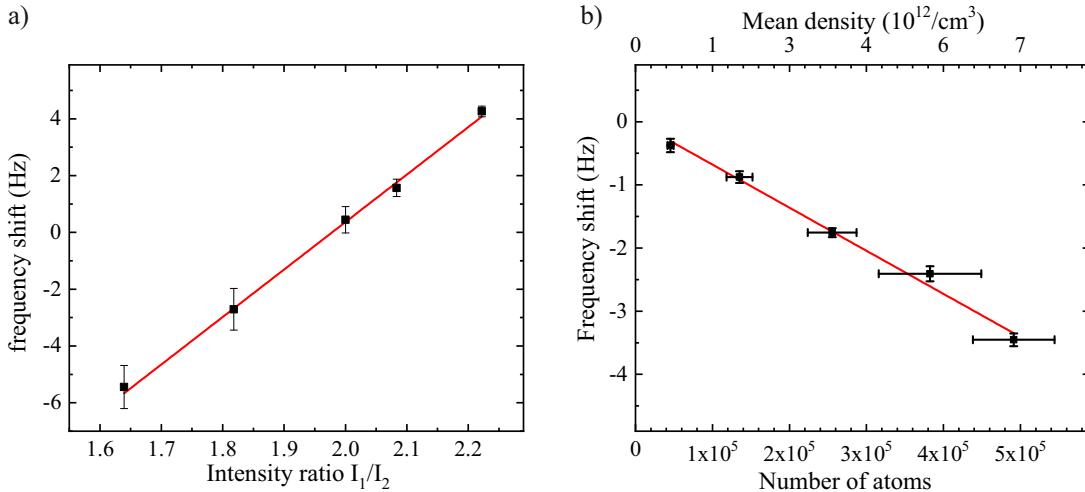


FIG. S2. (a) Frequency shift observed using the Raman transition relative to the frequency obtained by using the corresponding microwave transition. The shift vanishes for an intensity ratio of $I_1/I_2 = 1.98$. (b) Calibration of the mean density. The measured frequency of the carrier transition is shown as a function of the recorded number of atoms. Based on the known density-dependent frequency shift for Rb atoms [S2], the mean density of the ensemble is obtained.

The first-order motional sidebands can only be addressed with a coupling field that is asymmetric with respect to the trapping potential. This asymmetry is realized by shifting the Raman beams compared to the center position of the atomic cloud as shown in Fig. S1(b) and leads to a spatial inhomogeneity of the coupling.

The spectroscopic signals shown in of Fig. 1 and Fig. 2 (main text) are obtained by applying Raman pulses with a duration of 500 ms for detunings between ≈ -200 Hz and $\approx +200$ Hz of the two Raman laser beams. In Fig. 1 (main text), the carrier and all six sidebands are well resolved and, in addition, a higher order sideband is visible at ≈ 192 Hz. Compared to previous work [S4], the small Fourier width of these pulses allow for a full resolution of the sideband transitions.

At the end of each experimental sequence, the trap is switched off to allow for ballistic expansion and Stern-Gerlach separation of the atoms in the two clock states. The number of atoms in both states, N_\uparrow and N_\downarrow , and their temperature are detected by simultaneous absorption imaging, calibrated according to Ref. [S5].

S2. SPIN MODEL AND MEAN-FIELD DYNAMICS

In the main text we show that long-range XXZ spin models describe trapped bosonic gases interacting via purely contact interactions. Here we discuss the various parameters of the XXZ spin models and derive the corresponding mean-field equations of motion. We use them to calculate associated Rabi lineshapes.

Recall the Hamiltonian H_{XXZ} defined in main text,

$$H_{\text{XXZ}} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{ij} \chi_{ij} S_i^z S_j^z + \sum_i \Omega_i S_i^x - \sum_i (\delta - B_i) S_i^z. \quad (\text{S1})$$

This Hamiltonian describes the spin dynamics of thermal bosonic gases in the collisionless regime. By adequately mapping the harmonic trap eigenmodes to lattice sites in mode space, we can understand the spin dynamics for the carrier transition, the blue sideband transition, as well as the red sideband transition. The definitions of the two spin states in lattice site i for all these cases are as follows:

- Carrier transition: $|\uparrow\rangle_i = |\uparrow; n_i^X, n_i^Y, n_i^Z\rangle$, $|\downarrow\rangle_i = |\downarrow; n_i^X, n_i^Y, n_i^Z\rangle$
- Blue sideband transition (\hat{Z} direction): $|\uparrow\rangle_i = |\uparrow; n_i^X, n_i^Y, n_i^Z + 1\rangle$, $|\downarrow\rangle_i = |\downarrow; n_i^X, n_i^Y, n_i^Z\rangle$
- Red sideband transition (\hat{Z} direction): $|\uparrow\rangle_i = |\uparrow; n_i^X, n_i^Y, n_i^Z - 1\rangle$, $|\downarrow\rangle_i = |\downarrow; n_i^X, n_i^Y, n_i^Z\rangle$

Here we will use the convention of capital letters to denote spatial coordinates to distinguish them from coordinates in spin space denoted by lowercase letters. We denote the wave functions associated with $|\uparrow\rangle_i$ and $|\downarrow\rangle_i$ states respectively as $\phi_i^\uparrow(\mathbf{R})$ and $\phi_i^\downarrow(\mathbf{R})$. To avoid confusion, we define δ as the laser detuning from the carrier transition, and this

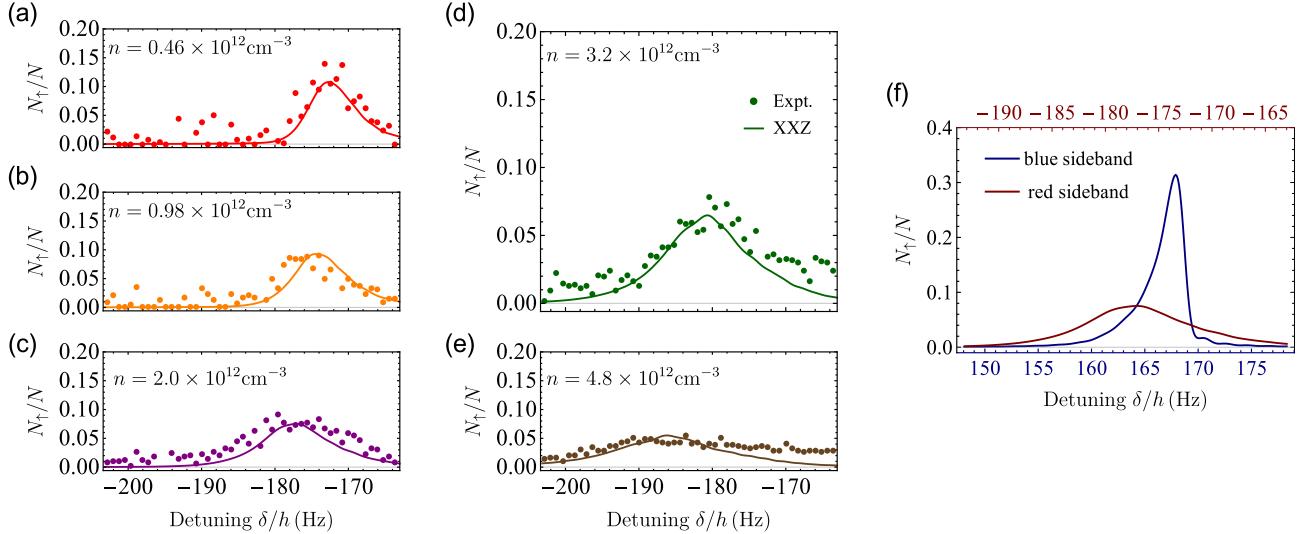


FIG. S3. (a-e) The Rabi lineshapes for 171 Hz red sideband with mean atom density $n = \{0.46, 0.98, 2.0, 3.2, 4.8\} \times 10^{12} \text{cm}^{-3}$. The solid points denote experimental data, the solid lines denote mean-field theoretical predictions by H_{XXZ} (see text). (f) The comparison of theoretical Rabi lineshapes between blue sideband and red sideband with mean density $n = 2.0 \times 10^{12} \text{cm}^{-3}$, and the difference can be understood as the effect of the anharmonicity of the optical dipole trap (see text).

convention is also used in the main text. For the blue sideband transition, we replace δ by $\delta - \hbar\omega$, where ω is the relevant trapping frequency; while for red sideband transition, we replace δ by $\delta + \hbar\omega$.

As we discuss in the main text, the key approximations in our spin model are the collisionless regime (trapping frequency is much larger than the interaction strength) as well as a negligible number of doubly occupied modes. Our experimental system is a thermal cloud of ^{87}Rb atoms prepared at a temperature $T = 375(25)$ nK in a 3D harmonic trap with trapping frequencies $\omega_X/2\pi = 143$ Hz, $\omega_Y/2\pi = 21.5$ Hz, $\omega_Z/2\pi = 171$ Hz. Based on the numerical calculation described below, we find $N\chi/h \approx 4.63 \text{Hz}/10^{12} \text{cm}^{-3}$ for blue sideband transition in the \hat{Z} direction, which demonstrates the validity of collisionless regime. For the number of doubly occupied modes, we evaluate the quantity $\sum_i \langle \hat{n}_i(\hat{n}_i - 1) \rangle / 2N$, where $\hat{n}_i = \hat{a}_i^\dagger \hat{a}_i$, and we are summing over all possible eigenmodes. This quantity is only non-zero when there is more than one atom per mode. Using Wick's theorem, we have

$$\frac{1}{2N} \sum_i \langle \hat{n}_i(\hat{n}_i - 1) \rangle = \frac{1}{2N} \sum_i \langle \hat{a}_i^\dagger \hat{a}_i^\dagger \hat{a}_i \hat{a}_i \rangle = \frac{1}{N} \sum_i \langle \hat{a}_i^\dagger \hat{a}_i \rangle \langle \hat{a}_i^\dagger \hat{a}_i \rangle = \frac{1}{N} \sum_i \langle \hat{n}_i \rangle^2 \quad (\text{S2})$$

Then we can estimate the average occupation number of each eigenmode by a Boltzmann distribution, $\langle \hat{n}_i \rangle / N = \exp[-(n^X \omega_X + n^Y \omega_Y + n^Z \omega_Z) \hbar / k_B T] / Z$, in which the partition function $Z \approx (k_B T / \hbar \bar{\omega})^3$. Then we get

$$\frac{1}{2N} \sum_i \langle \hat{n}_i(\hat{n}_i - 1) \rangle \approx N \left(\frac{\hbar \bar{\omega}}{2k_B T} \right)^3 \quad (\text{S3})$$

For $N = 1 \times 10^5$ atoms, we obtain that the number of occupied eigenmodes with two atoms or more is $N(\hbar \bar{\omega} / 2k_B T)^3 \sim 1.4\%$. This result leads to our conclusion that the number of lattice sites occupied by more than one atoms is very small in our experiment.

For the bosonic gas of interest, the Heisenberg couplings J_{ij} and the Ising couplings χ_{ij} are purely generated by contact interactions, and can be written in terms of the various scattering lengths and overlap integrals of harmonic trap eigenmodes,

$$J_{ij} = \frac{4\pi\hbar^2 a_{\uparrow\downarrow} V_{ij}^{\text{ex}}}{m}, \quad \chi_{ij} = \frac{4\pi\hbar^2 (V_{ij}^{\uparrow\uparrow} a_{\uparrow\uparrow} + V_{ij}^{\downarrow\downarrow} a_{\downarrow\downarrow} - V_{ij}^{\uparrow\downarrow} a_{\uparrow\downarrow} - V_{ij}^{\downarrow\uparrow} a_{\downarrow\uparrow})}{m}, \quad (\text{S4})$$

where

$$V_{ij}^{\alpha\beta} = \int d^3\mathbf{R} [\phi_i^\alpha(\mathbf{R})]^2 [\phi_j^\beta(\mathbf{R})]^2, \quad V_{ij}^{\text{ex}} = \int d^3\mathbf{R} \phi_i^\uparrow(\mathbf{R}) \phi_i^\downarrow(\mathbf{R}) \phi_j^\uparrow(\mathbf{R}) \phi_j^\downarrow(\mathbf{R}). \quad (\text{S5})$$

Besides spin-spin interaction, the Raman laser couples $|\uparrow_i\rangle$ and $|\downarrow_i\rangle$ states via a intermediate state, and the effective Rabi frequencies are given by

$$\Omega_i = \Omega_0 \int d^3\mathbf{R} \exp \left[-\frac{2(\mathbf{R}_\perp - \mathbf{R}_{\perp,0})^2}{w^2} \right] \phi_i^\uparrow(\mathbf{R}) \phi_i^\downarrow(\mathbf{R}). \quad (\text{S6})$$

Here, Ω_0 is the bare Rabi frequency determined by the Rabi couplings and detunings to the intermediate state of the two Raman beams, \mathbf{R}_\perp is perpendicular to the propagating direction of Raman beams, $\mathbf{R}_{\perp,0}$ is the offset from trap center, and w is the Gaussian beam radius at the position of atomic cloud. Due to the copropagating geometry of Raman beams used in the experiment, the momentum kicks of Raman beams can be ignored.

The different inhomogeneous longitudinal fields $B_i = B_i^{(1)} + B_i^{(2)} + B_i^{(3)}$ come from the interplay between contact interaction ($B_i^{(1)}$), the differential frequency shift generated by the optical trap and the magnetic curvature ($B_i^{(2)}$), and the anharmonicity in optical trap ($B_i^{(3)}$). We already discuss $B_i^{(1)}$ in the main text. It is given by

$$B_i^{(1)} = \frac{4\pi\hbar^2}{m} \sum_{j \neq i} (V_{ij}^{\uparrow\uparrow} a_{\uparrow\uparrow} - V_{ij}^{\downarrow\downarrow} a_{\downarrow\downarrow}). \quad (\text{S7})$$

The different trapping frequency experienced by the $|\uparrow\rangle$ and $|\downarrow\rangle$ atoms give an additional differential frequency shift in mode space. If we defined $\Delta\omega_{X,Y,Z} = \omega_{X,Y,Z}^\uparrow - \omega_{X,Y,Z}^\downarrow$, we can express $B_i^{(2)}$ as

$$B_i^{(2)} = \left(n_i^X + \frac{1}{2} \right) \hbar \Delta\omega_X + \left(n_i^Y + \frac{1}{2} \right) \hbar \Delta\omega_Y + \left(n_i^Z + \frac{1}{2} \right) \hbar \Delta\omega_Z. \quad (\text{S8})$$

Moreover, the actual Gaussian shape of the laser beams that make the dipole trap introduces corrections beyond the leading order harmonic trapping potential $U(\mathbf{R}) = \frac{1}{2}m(\omega_X^2 X^2 + \omega_Y^2 Y^2 + \omega_Z^2 Z^2)$. These corrections generate an additional anharmonic potential $\Delta U(\mathbf{R}) = -\frac{1}{2}m(\gamma_{XX}^2 X^4 + \gamma_{YY}^2 Y^4 + \gamma_{ZZ}^2 Z^4 + \gamma_{XY}^2 X^2 Y^2 + \gamma_{XZ}^2 X^2 Z^2 + \gamma_{YZ}^2 Y^2 Z^2)$. In first-order perturbation, this term gives rise to a shift of harmonic oscillator levels, which leads to a small change of atom density. As the shift generated by the anharmonicity is mode-dependent, it generates an extra longitudinal field that should be taken in consideration for sideband transitions. This field has a sign difference for the blue sideband ($B_i^{(3)b}$) and red sideband ($B_i^{(3)r}$). Here we use the sideband transitions in \hat{Z} direction as an example,

$$B_i^{(3)b} = -\frac{\hbar}{2\omega_Z} \left[3\gamma_{ZZ}^2 (a_{ho}^Z)^2 (n_i^Z + 1) + \gamma_{YZ}^2 (a_{ho}^Y)^2 (n_i^Y + 1/2) + \gamma_{XZ}^2 (a_{ho}^X)^2 (n_i^X + 1/2) \right], \quad (\text{S9})$$

$$B_i^{(3)r} = \frac{\hbar}{2\omega_Z} \left[3\gamma_{ZZ}^2 (a_{ho}^Z)^2 n_i^Z + \gamma_{YZ}^2 (a_{ho}^Y)^2 (n_i^Y + 1/2) + \gamma_{XZ}^2 (a_{ho}^X)^2 (n_i^X + 1/2) \right], \quad (\text{S10})$$

where $a_{ho}^{X,Y,Z} = \sqrt{\hbar/m\omega_{X,Y,Z}}$ is the harmonic oscillator length. Sideband transitions in other directions can be treated in a similar way.

For carrier transition, due to the negligible Ising couplings χ_{ij} , our XXZ spin model can be simplified to the Heisenberg model. Given that the transverse field Ω_i and longitudinal field B_i are small compared to the Heisenberg couplings J_{ij} , we can restrict the spin model in the Dicke manifold, which gives $H_{\text{carrier}} \approx \Omega S^x - (\delta - B)S^z$, where Ω is the mean Rabi frequency for carrier transition, and B is the thermal-averaged value of B_i . We understand B as the frequency shift of carrier transition, which can be evaluated analytically in the large- N limit,

$$B = \frac{4\pi\hbar^2(a_{\uparrow\uparrow} - a_{\downarrow\downarrow})n}{m} + k_B T \left(\frac{\Delta\omega_X}{\omega_X} + \frac{\Delta\omega_Y}{\omega_Y} + \frac{\Delta\omega_Z}{\omega_Z} \right), \quad (\text{S11})$$

where $n = N(m\bar{\omega}^2/4\pi k_B T)^{3/2}$ is the mean atom density in harmonic trap with atom number N , and $\bar{\omega} = (\omega_X \omega_Y \omega_Z)^{1/3}$. The density-dependent part in B agrees with the density-dependent clock shift $-0.48 \text{ Hz}/10^{12} \text{ cm}^{-3}$ observed in previous experiment [S2]. In our experiment, we use this known value of density-dependent shift of the carrier transition to calibrate the atom density.

For sideband transitions, the Ising couplings χ_{ij} become larger. We use a mean-field approximation, which neglects the quantum correlation between different spins, $\langle S_i^\mu S_j^{\mu'} \rangle \approx \langle S_i^\mu \rangle \langle S_j^{\mu'} \rangle$ ($\mu, \mu' = x, y, z$), to derive Heisenberg equations

of $S_i^{x,y,z}$ in our XXZ spin model (see Eq.(S1)). The mean-field equations we get are the following ones:

$$\begin{aligned}\frac{d}{dt}\langle S_j^x \rangle &= 2 \sum_i \left[J_{ij} \langle S_i^y \rangle \langle S_j^z \rangle - (J_{ij} + \chi_{ij}) \langle S_i^z \rangle \langle S_j^y \rangle \right] + (\delta - B_j) \langle S_j^y \rangle, \\ \frac{d}{dt} S_j^y &= 2 \sum_i \left[(J_{ij} + \chi_{ij}) \langle S_i^z \rangle \langle S_j^x \rangle - J_{ij} \langle S_i^x \rangle \langle S_j^z \rangle \right] - (\delta - B_j) \langle S_j^x \rangle - \Omega_j \langle S_j^z \rangle, \\ \frac{d}{dt} \langle S_j^z \rangle &= 2 \sum_i J_{ij} \left[\langle S_i^x \rangle \langle S_j^y \rangle - \langle S_i^y \rangle \langle S_j^x \rangle \right] + \Omega_j \langle S_j^y \rangle.\end{aligned}\quad (\text{S12})$$

We solve Eq.(S12) numerically, with random sampling of motional states drawn from a Boltzmann distribution. As it is computationally difficult to solve the equations above for $\sim 10^5$ atoms, instead we use $N_{\text{th}} = 1000$ and scale the transverse and longitudinal field from the one in the experiment by a factor $N_{\text{th}}/N_{\text{exp}}$. We also allow an overall scaling factor η of the atomic density to take both finite-size effects and the anharmonicities into account. The thermal-averaged sideband spectrum agrees well with our experimental measurements, when the overall scaling factor is set to $\eta = 0.72$ for both blue sideband and red sideband. The Rabi spectrum of blue sideband transition is discussed in the main text (see Fig. 3(a-c,e-f)), and the Rabi spectrum of red sideband transition is depicted in Fig. S3(a-e). We also compare the theoretical Rabi lineshapes for blue and red sideband for mean atom density $n = 2.0 \times 10^{12} \text{ cm}^{-3}$ in Fig. S3(f), which shows a significant suppression of red sideband. As the temperature of our system is above quantum degeneracy, the ground state concentration is not a reasonable explanation. Instead, the difference between the blue and red sidebands comes from a sign difference between $B_i^{(3)b}$ and $B_i^{(3)r}$, generated by anharmonicity. Because of this sign difference, $B_i^{(3)b}$ partially cancels the inhomogeneity in the longitudinal fields, while $B_i^{(3)r}$ increases the inhomogeneity. Similar phenomenon was also observed in Ref. [S6].

S3. DYNAMICAL PHASE DIAGRAM AND CRITICAL BEHAVIOR

In the main text we discuss about the ferromagnetic to paramagnetic dynamical phase transition (DPT) in the Lipkin-Meshkov-Glick (LMG) model (see Eq.(3) main text), a collective XXZ model plus additional transverse and longitudinal field. Here we elaborate on the calculation of the dynamical phase diagram and the associated critical points, following the procedure discussed in Ref. [S7]. The mean-field equations of the LMG model can be written in terms of normalized expectation value of total spin operators $s^{x,y,z} = 2\langle S^{x,y,z} \rangle/N$ as follows,

$$\begin{aligned}\frac{d}{dt} s^x &= -N\chi s^z s^y + \tilde{\delta} s^y, \\ \frac{d}{dt} s^y &= N\chi s^z s^x - \tilde{\delta} s^x - \Omega s^z, \\ \frac{d}{dt} s^z &= \Omega s^y.\end{aligned}\quad (\text{S13})$$

Using both energy conservation in H_{LMG} , for an initial state with $s^z = -1$, $s^x = s^y = 0$, as well as the identity $(S^x)^2 + (S^y)^2 + (S^z)^2 = (\frac{N}{2} + 1)\frac{N}{2}$ in large- N limit, the mean-field variables satisfy the following two conservation relations:

$$\frac{N\chi}{2} s^z s^z - \tilde{\delta} s^z + \Omega s_x = \frac{N\chi}{2} + \tilde{\delta}, \quad (\text{S14})$$

$$(s^x)^2 + (s^y)^2 + (s^z)^2 = 1. \quad (\text{S15})$$

Combining these three equations (Eq.(S13)-(S15)), we can eliminate s^x and s^y , and obtain the following differential equation for s^z ,

$$\frac{1}{2} \left(\frac{d}{dt} s^z \right)^2 + V(s^z) = 0, \quad (\text{S16})$$

where

$$V(s^z) = (s^z + 1) \left\{ \frac{(N\chi)^2}{8} (s^z)^3 - \left[\frac{(N\chi)^2}{8} + \frac{N\chi\tilde{\delta}}{2} \right] (s^z)^2 + \left[\frac{\tilde{\delta}^2 + \Omega^2}{2} - \frac{(N\chi)^2}{8} \right] s^z + \left[\frac{\tilde{\delta}^2 - \Omega^2}{2} + \frac{N\chi\tilde{\delta}}{2} + \frac{(N\chi)^2}{8} \right] \right\}. \quad (\text{S17})$$

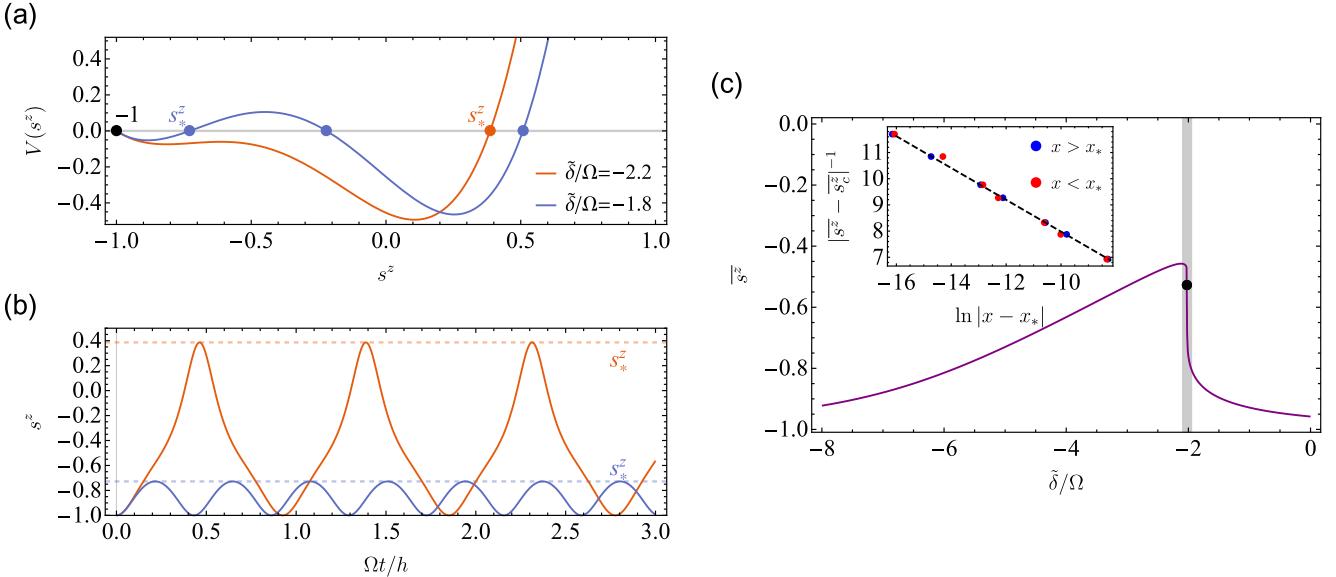


FIG. S4. (a) The effective potential $V(s^z)$ with $N\chi/\Omega = 5$. In the case of $\tilde{\delta}/\Omega = -2.2$, $V(s^z)$ has two real roots; In the case of $\tilde{\delta}/\Omega = -1.8$, $V(s^z)$ has four real roots. The nearest turnover point is labelled by s_*^z , and the jump of s_*^z indicates dynamical phase transition (see text). (b) The mean-field dynamics of LMG model with $N\chi/\Omega = 5$ and $\tilde{\delta}/\Omega = -2.2, -1.8$, which shows a sharp change of mean-field dynamical behavior. The choice of color for the lines is the same as (a), and the dashed lines mark the nearest turnover point s_*^z . (c) The long-time average value \bar{s}^z with $N\chi/\Omega = 5$, and the critical point is labelled by black circle. The shaded area is the region close to the critical point, which is shown in details in the inset with logarithmic non-analyticity at the critical point (see text).

We interpret Eq.(S16) as the Hamiltonian of a classical particle with position s^z moving in the effective potential $V(s^z)$. The condition $V(s^z) = 0$ determines the turning points of s^z . Since $V(-1) = 0$, $V'(-1) = -1$, $V(1) = \tilde{\delta}^2$, this effective potential should have at least two real roots in $[-1, 1]$, and we consider these roots as physical turning points. So the dynamics of s^z can be understood as the oscillations between -1 and the nearest turnover point s_*^z . Imagine that we start from a $V(s^z)$ with two real roots, and continuously tune the parameters of $V(s^z)$ so that two new real roots appear in between, a jump of the nearest turning point s_*^z should occur in this process (see Fig. S4(a-b)). This abrupt change in behavior is what sets the dynamical phase transition.

To count the number of roots in $V(s^z)$, we can factor out the known root $s^z = -1$, and then consider the discriminant $\Delta = 18abcd - 4b^3d + b^2c^2 - 4ac^3 - 27a^2d^2$ of cubic equation $ax^3 + bx^2 + cx + d = 0$. If $\Delta > 0$, the cubic has three distinct real roots; if $\Delta < 0$, the cubic has one real root. So $\Delta = 0$ captures the critical point of the DPT. We focus on the parameter regime where $N\chi > 0$ with a fixed positive Ω , and define $y = N\chi/\Omega$, $x = \tilde{\delta}/\Omega$. In terms of these variables the phase boundary plotted in the main text (see Fig. 2(d)) is given by

$$y_* = \frac{1}{12x_*} \left[1 - 12x_*^2 - \left(5832x_*^4 + 540x_*^2 - 1 + 24x_*\sqrt{3(27x_*^2 - 1)^3} \right)^{1/3} - \left(5832x_*^4 + 540x_*^2 - 1 - 24x_*\sqrt{3(27x_*^2 - 1)^3} \right)^{1/3} \right]. \quad (\text{S18})$$

As this formula includes square root and cube root, we need to specify the argument of complex number to avoid the branch cut. Here we choose $\arg[3(27x_*^2 - 1)^3] = \{0, \pi\}$, $\arg[5832x_*^4 + 540x_*^2 - 1 + 24x_*\sqrt{3(27x_*^2 - 1)^3}] \in (-2\pi, 0]$, and $\arg[5832x_*^4 + 540x_*^2 - 1 - 24x_*\sqrt{3(27x_*^2 - 1)^3}] \in [0, 2\pi]$. And we can conclude that this phase boundary exists in the regime $x < \sqrt{3}/9$ and $y > 8\sqrt{3}/9$. Therefore, only when $N\chi/\Omega > 8\sqrt{3}/9$, the DPT occurs. Instead, if $N\chi/\Omega < 8\sqrt{3}/9$, there is a smooth crossover.

As we mentioned in the main text, our experiment always lie in the DPT regime, and we characterize the ferromagnetic and paramagnetic phase using the long-time average of excitation fraction \bar{N}_\uparrow/N , which is possible to express in terms of s^z ,

$$\frac{\bar{N}_\uparrow}{N} = \frac{1}{2}(\bar{s}^z + 1), \quad \bar{s}^z = \frac{1}{T} \int_0^T s^z(t) dt, \quad (\text{S19})$$

where T is the oscillation period of s^z . This integral can be evaluated using Eq.(S16), as

$$\int_0^T s^z(t) dt = \int_{-1}^{s_*^z} \frac{2s^z ds^z}{\sqrt{-2V(s^z)}}, \quad T = \int_{-1}^{s_*^z} \frac{2 ds^z}{\sqrt{-2V(s^z)}}. \quad (\text{S20})$$

All these integrals can be calculated analytically in terms of elliptic integrals. Considering the asymptotic behavior near the critical point, we find that instead of the sudden jump behavior of s_*^z , the long-time average \bar{s}^z is continuous at the critical point, and the first derivative of \bar{s}^z diverge logarithmically. The following formula describe the asymptotic behavior of \bar{s}^z with fixed $N\chi/\Omega$,

$$\bar{s}^z \rightarrow \bar{s}_c^z + \frac{C}{\ln|x - x_*|}, \quad \bar{s}_c^z = \frac{1}{2} - \frac{1}{2} \sqrt{1 - \frac{8x_*}{y_*}}, \quad (\text{S21})$$

where C is a constant set by $N\chi/\Omega$. We can verify the asymptotic behavior predicted above numerically. This is plotted in Fig. S4(c) for the case $N\chi/\Omega = 5$. The continuous behavior of s_*^z at the critical point determines a second-order dynamical phase transition in our case.

Here we also discuss how to determine the critical point in experiment. Based on Fig. S4(c), the first derivative of the long-time average excitation fraction diverges at the critical point. However, in the analysis of experimental data, we have to use the finite difference as an approximation of the first derivative, which is limited by the precision of laser frequency and experimental fluctuation. To construct a stable phase boundary, instead we use the maximum transfer point as a signature of the critical point. Also, as we mentioned in the main text, we measure the Rabi spectrum at a fixed time instead of taking the long-time average excitation fraction due to technical challenges. All these systematic errors in determination of the critical point are smaller than the measurement error bars under current experimental conditions.

Finally we discuss the scaling factor in χ used in the main text to match the experimental critical points to the DPT in LMG model. This scaling factor originates from the inhomogeneities in the Ising couplings χ_{ij} , which couple the Dicke manifold to the states with different total spins. In this way, the effective Ising coupling should be modified by an overall factor from its value χ in Dicke manifold. We use the same scaling factor 0.56 for all the measurements with different atom densities, and the experimental critical points agrees very well with the phase boundary in LMG model.

S4. DISCUSSION OF SPIN SQUEEZING

We proceed to study the role of quantum correlations and entanglement in our XXZ simulator by theoretical calculation of spin squeezing, since it provides a relevant entanglement witness and an important resource for quantum metrology [S8]. We study the proposed Ramsey spectroscopy sequence depicted in Fig. S5(a). Initially all atoms are assumed to be in the $|\downarrow\rangle$ state and a $\pi/2$ blue-sideband pulse is applied to transfer the atoms to the $|S^x = N/2\rangle$ state. Then the system is allowed to evolve for $\tau/2$ under XXZ interaction (see Eq.(2) in the main text), followed by a blue-sideband spin echo pulse, and a further evolution time $\tau/2$. The additional spin echo pulse at half of the evolution suppresses the dephasing effect of inhomogeneous longitudinal fields. The squeezing is quantified by the Ramsey spin squeezing parameter [S9], $\xi^2 = \min_\theta N(\Delta S_\theta^\perp)^2 / |\langle \mathbf{S} \rangle|^2$, which signals entanglement if $\xi^2 < 1$. Here, $(\Delta S_\theta^\perp)^2$ is the variance of the spin noise along an axis perpendicular to the collective spin $\langle \mathbf{S} \rangle$, parametrized by an angle $\theta \in [0, 2\pi]$. This squeezing parameter can be extracted by an appropriate sequence of spin rotations at the end of the Ramsey protocol.

To estimate the achievable spin squeezing, we adopt the discrete truncated Wigner approximation (DTWA), which solves the mean-field equations of motion supplemented by Monte Carlo sampling of the initial conditions to account for quantum fluctuations [S10]. We choose $N_{\text{th}} = 1000$ and scale the longitudinal field from the one in the experiment by a factor $N_{\text{th}}/N_{\text{exp}}$. The theoretical prediction of spin squeezing is depicted in Fig. S5(b). We compare it to the spin squeezing in the pure XXZ model (ignoring longitudinal fields in Eq.(2) in the main text), and the Ising model ($H_{\text{Ising}} = \sum_{ij} \chi_{ij} S_i^z S_j^z$) which allows for an exact solution (see below). We find that the beyond-mean-field dynamics in our simulator is similar to the Ising limit, with an additional small suppression arising from the inhomogeneities in the longitudinal fields. For $N_{\text{th}} = 1000$ atoms, near 6dB optimal squeezing can be achieved at $N\chi\tau/h \approx 0.5$, which translates under current experimental conditions to optimal squeezing times around 100ms. On this time scale we do not expect detrimental effects from the technical imperfections. The predicted squeezing emphasizes the metrological potential of motional sidebands.

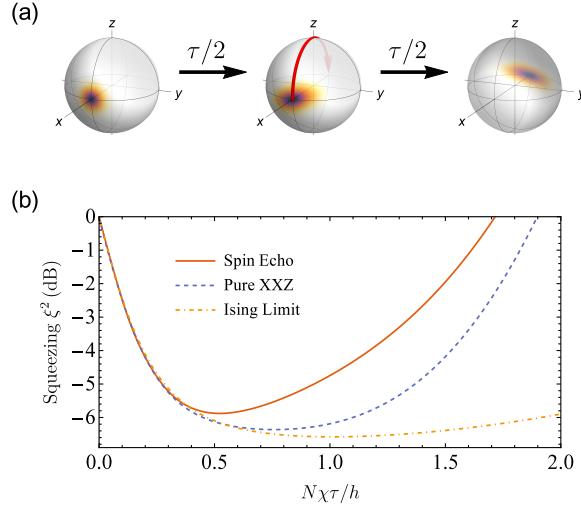


FIG. S5. (a) Spin echo sequence for generation of squeezing using motional sidebands (see text). The total spin state $|\Psi(t)\rangle$ is illustrated using the Husimi-Q function [S8]. (b) Comparison of the obtained spin squeezing for a spin echo sequence (DTWA method), the pure XXZ model (DTWA method) and the Ising limit (analytic solution). The spin squeezing parameter is expressed in terms of decibels (dB), i.e. $10 \log_{10} \xi^2$.

A. Analytic solution for Ising model

In Fig. S5, we compare the theoretical calculation of the achievable spin squeezing in our XXZ simulator with the analytic solution of Ising model ($\hbar = 1$),

$$H_{\text{Ising}} = \sum_{ij} \chi_{ij} S_i^z S_j^z. \quad (\text{S22})$$

Without loss of generality, we assume the Ising couplings are symmetric, $\chi_{ij} = \chi_{ji}$. As we start from the initial state $|S^x = N/2\rangle$, it is easy to show that the collective spin always stays in x direction, and we can simplify the definition of spin squeezing parameter ξ^2 in the main text as follow,

$$\xi^2 = \min_{\theta} \frac{N(\Delta S_{\theta}^{\perp})^2}{|\langle S^x \rangle|^2}, \quad (\text{S23})$$

where

$$(\Delta S_{\theta}^{\perp})^2 = \cos^2 \theta \langle S^z S^z \rangle + \sin^2 \theta \langle S^y S^y \rangle + \cos \theta \sin \theta \langle S^z S^y + S^y S^z \rangle. \quad (\text{S24})$$

Following the technique of discussing one-axis-twisting model in Ref. [S8], all the expectation values above can be evaluated as follows,

$$\begin{aligned} \langle S^x \rangle &= \frac{1}{2} \sum_k \prod_j^{(k)} \cos(\chi_{kj} t), \quad \langle S^z S^z \rangle = \frac{N}{4}, \\ \langle S^y S^y \rangle &= \frac{N}{4} + \frac{1}{4} \sum_{k < l} \left[\prod_j^{(k,l)} \cos[(\chi_{kj} - \chi_{lj})t] - \prod_j^{(k,l)} \cos[(\chi_{kj} + \chi_{lj})t] \right], \\ \langle S^z S^y + S^y S^z \rangle &= \frac{1}{2} \sum_{k < l} \sin(\chi_{kl} t) \left[\prod_j^{(k,l)} \cos(\chi_{kj} t) + \prod_j^{(k,l)} \cos(\chi_{lj} t) \right], \end{aligned} \quad (\text{S25})$$

where $\prod_j^{(k)}$ means multiplication without the term $j = k$. Based on all these expectation values, we can calculate the spin squeezing parameter ξ^2 by tuning θ to reach the minimum value of $(\Delta S_{\theta}^{\perp})^2$. We define this angle as optimal

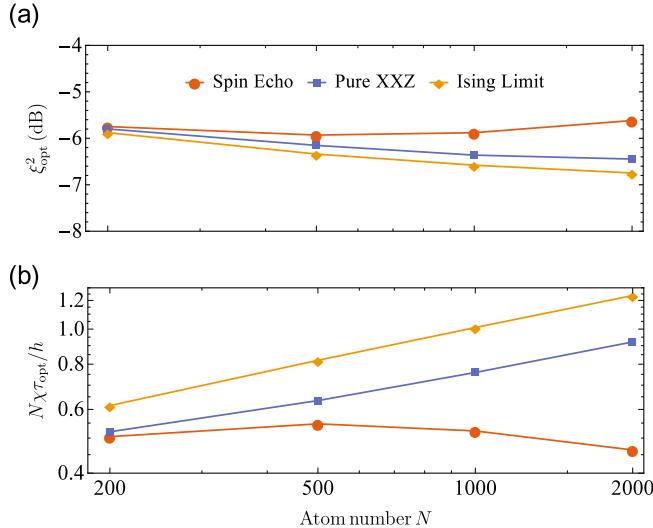


FIG. S6. (a) Finite-size scaling of optimal spin squeezing in our XXZ simulator with spin echo sequence (DTWA method), compared to pure XXZ model (DTWA method) and Ising limit (analytic solution). (b) Finite-size scaling of optimal squeezed time in our XXZ simulator with spin echo sequence (DTWA method), compared to pure XXZ model (DTWA method) and Ising limit (analytic solution).

squeezed angle θ_0 , and we get

$$\begin{aligned} \min_{\theta} (\Delta S_{\theta}^{\perp})^2 &= \frac{A+B}{2} - \frac{1}{2}\sqrt{(B-A)^2 + C^2} \\ \tan(2\theta_0) &= \frac{-C}{B-A} \end{aligned} \quad (\text{S26})$$

where $A = \langle S^z S^z \rangle$, $B = \langle S^y S^y \rangle$, $C = \langle S^z S^y + S^y S^z \rangle$. In this way, the spin squeezing for general case of Ising interaction can be evaluated analytically.

B. Prediction of achievable spin squeezing

In Fig. S5, we discuss the achievable spin squeezing in our XXZ simulator, with comparison to the pure XXZ model and the Ising limit. As it is hard to calculate spin squeezing for $\sim 10^5$ atoms in theory, we choose $N_{\text{th}} = 1000$ and scale the longitudinal field from the one in the experiment by a factor $N_{\text{th}}/N_{\text{exp}}$. The spin squeezing as a function of Ramsey dark time is depicted in the main text (see Fig. 4(b)). Here we use finite-size scaling as a way to predict the achievable spin squeezing under experimental conditions (see Fig. S5).

We extract the optimal spin squeezing (see Fig. S5(a)) and optimal squeezed time (see Fig. S5(b)) with $N_{\text{th}} = 200, 500, 1000, 2000$. We find that under current experimental conditions, the optimal squeezing saturates around 6dB when we increase the atom number in theory, and the optimal squeezed time stays near $N\chi\tau/h \approx 0.5$. Unfortunately the finite-size scaling curve for the current experiment condition is not monotonic, which means that our estimation of the optimal spin squeezing is not necessarily accurate. The analysis nevertheless shows the detrimental effects caused by the inhomogeneities in longitudinal fields, which will lead to a non-negligible suppression of spin squeezing, compared to pure XXZ model and Ising limit. Therefore we predict that if it were possible to carefully control the longitudinal fields in experiment and reduce their size, one could get closer to the finite-size scaling curve of pure XXZ model, which increases monotonically when increasing atom number, although the improvement is not significant due to the thermal distribution. In this case, we predict an optimal spin squeezing set by $\xi_{\text{opt}}^2 \propto N^{-0.067}$, and an optimal squeezed time by $\tau_{\text{opt}} \propto N^{-0.752}$. Ideally speaking, for 5×10^5 atoms, 8dB optimal squeezing can be achieved around 100ms.

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- [S1] Y.-J. Lin, A. R. Perry, R. L. Compton, I. B. Spielman, and J. V. Porto, *Phys. Rev. A* **79**, 063631 (2009).
 - [S2] G. Kleine Bünning, J. Will, W. Ertmer, E. Rasel, J. Arlt, C. Klempt, F. Ramirez-Martinez, F. Piéchon, and P. Rosenbusch, *Phys. Rev. Lett.* **106**, 240801 (2011).
 - [S3] A. Kaplan, M. F. Andersen, and N. Davidson, *Phys. Rev. A* **66**, 045401 (2002).
 - [S4] W. Maineult, C. Deutsch, K. Gibble, J. Reichel, and P. Rosenbusch, *Phys. Rev. Lett.* **109**, 020407 (2012).
 - [S5] G. Reinaudi, T. Lahaye, Z. Wang, and D. Guéry-Odelin, *Opt. Lett.* **32**, 3143 (2007).
 - [S6] B. Allard, M. Fadel, R. Schmied, and P. Treutlein, *Phys. Rev. A* **93**, 043624 (2016).
 - [S7] J. A. Muniz, D. Barberena, R. J. Lewis-Swan, D. J. Young, J. R. K. Cline, A. M. Rey, and J. K. Thompson, *Nature* **580**, 602 (2020).
 - [S8] J. Ma, X. Wang, C.-P. Sun, and F. Nori, *Phys. Rep.* **509**, 89 (2011).
 - [S9] D. J. Wineland, J. J. Bollinger, W. M. Itano, F. L. Moore, and D. J. Heinzen, *Phys. Rev. A* **46**, R6797 (1992).
 - [S10] J. Schachenmayer, A. Pikovski, and A. M. Rey, *Phys. Rev. X* **5**, 011022 (2015).