

x -polarized state, we probe the relaxation for initial states with different magnetization. The experimental protocol is shown in Fig. 9(a) and is similar to the one used in [53]. It consists of the following steps. In the preparation, spins are initially polarized along the x axis. A locking field Ω_{Lock} , which is also aligned along the x axis is applied for a time t_1 . During this time, as reported in [53], the magnetization will relax and approximately settle to a constant nonzero value that depends on the strength of the locking field. In the evolution, we then turn off the locking field and measure the resulting relaxation of the x -magnetization. The resulting relaxation over period t_2 is shown in Fig. 9(b). The magnetization starts with different values depending on the field strength applied during the preparation. We note that the locking time $t_2 = 2 \mu\text{s}$ is larger than the time it takes to directly relax to zero magnetization without phase one (blue points). We observed that for decreasing initial magnetization, the onset of the relaxation dynamics gets shifted to a later time (red, green, and yellow points). However, independent of this behavior, for later times, all curves overlap with the direct relaxation curve without field (blue points).

The observed dynamics can be understood within the pair approximation in the following way. During preparation, the locking field is only able to lock pairs with interactions smaller than the field strength Ω_{Lock} . These pairs stay polarized while pairs with stronger interactions oscillate and dephase. As reported in [53], magnetization takes an almost constant value. During the evolution, when the field is turned

off, the relaxation timescale is given by the remaining pairs that were locked and now start to oscillate. This timescale is longer for small fields where only weakly interacting pairs remained locked during the preparation. The overlapping at a later time is due to the fact that these pairs are also locked under larger fields. The data was taken for $|48S_{1/2}, m_j = 0.5\rangle$ and $|48P_{3/2}, m_j = 0.5\rangle$.

APPENDIX F: DERIVATION OF DEPOLARIZATION DYNAMICS

The goal is to compute the expectation value of $\langle \hat{S}_x(t) \rangle = \frac{1}{N} \sum_i \langle \hat{S}_x^i \rangle$ starting from the x -polarized state $|\psi_0\rangle = |\rightarrow\rangle^{\otimes N}$ governed by the effective Hamiltonian derived in [48]

$$\hat{H}_{\text{eff}} = \sum_{\langle i,j \rangle} (J_{ij}^{\perp} (\hat{S}_x^i \hat{S}_x^j + \hat{S}_y^i \hat{S}_y^j) + J_{ij}^{\parallel} \hat{S}_z^i \hat{S}_z^j) + \sum_{\langle i,j \rangle, \langle k,l \rangle} J_{\text{eff}}^{ijkl} \hat{S}_z^{(i)(j)} \hat{S}_z^{(k)(l)} \quad (\text{F1})$$

where $\langle i, j \rangle$ denotes the summation over paired spins i and j and $2\hat{S}_z^{(i)(j)} = \hat{S}_z^{(i)} + \hat{S}_z^{(j)}$.

Without loss of generality, we assume that spins 1 and 2 form a pair and compute $\langle \hat{S}_x^1(t) \rangle$. The evolution of $\langle \hat{S}_x(t) \rangle$ then follows simply by linearity. First we notice that all the terms in \hat{H}_{eff} commute with each other, allowing for direct computation of $\langle \hat{S}_x^1(t) \rangle$ by commuting \hat{S}_x^1 through the time evolution operators,

$$\hat{S}_x^1(t) = e^{it\hat{H}_{\text{eff}}} \hat{S}_x^1 e^{-it\hat{H}_{\text{eff}}} \quad (\text{F2})$$

$$= e^{itJ_{12}^{\perp} (\hat{S}_x^1 \hat{S}_x^2 + \hat{S}_y^1 \hat{S}_y^2)} e^{itJ_{12}^{\parallel} \hat{S}_z^1 \hat{S}_z^2} e^{it\hat{S}_z^{(1)(2)} \sum_{\langle k,l \rangle} J_{\text{eff}}^{12kl} \hat{S}_z^{(k)(l)}} \hat{S}_x^1 e^{-it\hat{S}_z^{(1)(2)} \sum_{\langle k,l \rangle} J_{\text{eff}}^{12kl} \hat{S}_z^{(k)(l)}} e^{itJ_{12}^{\perp} (\hat{S}_x^1 \hat{S}_x^2 + \hat{S}_y^1 \hat{S}_y^2)} e^{-itJ_{12}^{\parallel} \hat{S}_z^1 \hat{S}_z^2} \quad (\text{F3})$$

$$= e^{itJ_{12}^{\perp} (\hat{S}_x^1 \hat{S}_x^2 + \hat{S}_y^1 \hat{S}_y^2)} e^{it\hat{S}_z^1 \sum_{\langle k,l \rangle} J_{\text{eff}}^{12kl} \hat{S}_z^{(k)(l)}} e^{2itJ_{12}^{\parallel} \hat{S}_z^1 \hat{S}_z^2} e^{-itJ_{12}^{\perp} (\hat{S}_x^1 \hat{S}_x^2 + \hat{S}_y^1 \hat{S}_y^2)} \hat{S}_x^1. \quad (\text{F4})$$

Now we can just expand the exponentials using the usual formula for the exponential of Pauli matrices (note that $\hat{S}_x^1 \hat{S}_x^2 + \hat{S}_y^1 \hat{S}_y^2$ is akin to \hat{S}_x in a specific subspace) and take the expectation value with respect to the initial state to get the desired result,

$$\langle \hat{S}_x^1(t) \rangle = \frac{1}{2} \cos \left(\frac{J_{12}^{\perp} - J_{12}^{\parallel}}{2} t \right) \prod_{\langle k,l \rangle} \cos^2 \left(\frac{J_{\text{eff}}^{12kl}}{8} t \right). \quad (\text{F5})$$

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