

Large Spin Atoms

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Paris North University Villetaneuse

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Investigating quantum magnetism with large spin (s>1/2) particles

Our two experimental platforms at LPL

Investigating quantum magnetism with large spin (s>1/2) particles:

A Chromium **Bose-Einstein** condensate in a 3D optical lattice

Spin-dependent dipolar interactions S=3, 7 spin states

A SU(10) Strontium Fermi gas in a 3D optical lattice

Spin-independent contact interactions F=9/2, 10 spin states





... using collective variables ...

How to characterize a quantum many-body system

Density matrix ρ

$$\begin{bmatrix} \rho_{11} & \cdots & \rho_{1k} \\ \vdots & \ddots & \vdots \\ \rho_{1k} & \cdots & \rho_{kk} \end{bmatrix} \quad \oint \mathbf{k} \sim 2^{\mathbf{N}}$$

Full state tomography impractical

Partial information : trace over subsystem

Partial information : collective measurement

How to characterize a quantum many-body system

Partial information : trace over subsystem

Example for a pure state:

Tr(ho^2)=1

 $\rho = |\Psi\rangle\langle\Psi|$

« pure »



« locally mixed »

Measures the **entropy associated to entanglement**

See Greiner 2016 Klempt; Treutlein; Oberthaler 2018

(or more simply measure correlations between two sub-systems)

How to characterize a quantum many-body system

 $\sum_{i=1}^{N} \hat{s}_{z,i}$

Partial information : collective measurement

Entanglement witness

(e.g.
$$(\Delta S_x)^2 + (\Delta S_y)^2 + (\Delta S_z)^2 \ge N/2$$
 for any mixture of separable states)

Squeezing (Ueda)

Extreme spin squeezing (Sorensen/Molmer - Klempt) or Fischer information (Oberthaler):

 \rightarrow k-particle entanglement



Meets quantum metrology Connects to quantum optics

!!! Needs access to coherences **!!!**

!!! Beware of large spin systems !!!
(i.e. squeezing is not an EW) – See G. Toth

Fluctuations give access to correlations (here for S>1/2)



PART I : Strontium

Introduction to alkaline-earth atoms



Zero electronic spin: no magnetic field sensisivity



(see Takahashi)

How to manipulate spin?

Purely nuclear spin \rightarrow magnetic field inconvenient

Create an articificial magnetic field (spin-orbit coupling) (spindependent AC-Stark shift)

Use the intercombination line (low scattering rate, large hyperfine structure, large Landé factor)

Example : optical Stern-Gerlach to measure spin





Image on the broad 461 nm transition





NB ~anti SWAP cooling

Spin-dependent momentum transfert with an adiabatic sweep





Measuring the spin (one shot)



Three pulse

sequence



Diffracted fraction (Right)







PRELIMINARY



Transfer efficiency ~80%, limited by amplified ASE from laser diode



PART II : Chromium



Dipolar systems in ultra-cold atoms and molecules



dipolar molecules VS VS а $|1,0\rangle$ 2.2 GHz 0,0 difference b 1.0 Contrast 10 20 T (ms) 30 0.5 **Boulder** 0.0 20 50 10 30 40 0 T (ms)

transport possible; truly macroscopic Large spin Individual adressing coming up (Greiner)

Control of Hamiltonian Individual adressing coming up (Bakr) Control of Hamiltonian And geometry individual adressing



Rydberg Atoms



Quantum Thermalization (Isolated System)



Eigenstate Thermalization HypothesisDeutsch, Srednicki, Olshanii**Growth of entanglement**Deutsch, Srednicki, Olshanii

This talk : use of collective measurements (e.g.

g.
$$\sum_{i=1}^{N} \hat{s}_{z,i}$$
)

Outline:

1- Thermalization of the Zeeman populations

2- Thermalization of the collective spin

3- Experimental measurement of correlations using collective measurements

Experimental results



Agreement with GDTWA indicate the importance of quantum correlations



Fractionnal population

Asymptotic behavior



Take into account energy constraints.

Two contributions for energy

Dipole-dipole interactions

 $\langle \Psi(t)|V_{dd}|\Psi(t)\rangle$

Difficult to calculate except at t=0

Tensor light-shift leads to an effective quadratic Zeeman effect

$$E(m_s) = B_Q m_s^2$$

Simple to evaluate using experimental data

$$\sum m_s^2 p_{m_s}$$

Analytic model for quantum thermalization

Look at the thermal state that corresponds to the initial energy

High-temperature expansion (A.M. Rey)

Consistent with the eigenstate thermalization hypothesis (~the thermal character is built in the eigenstates themselves)



An effective temperature (a few nK) for an isolated (somewhat pure) system

Outline:

1- Thermalization of the Zeeman populations

2- Thermalization of the collective spin

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Measuring the collective spin through Ramsey interferometry

8

 $\ell = \sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2 + \langle S_z \rangle^2}$







Measuring the collective spin through Ramsey interferometry $\ell = \sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2 + \langle S_z \rangle^2}$



Damping of the collective spin due to dipolar interactions

Good agreement at short times Good agreement with second-order perturbation theory too

Hazzard et al., PRL 110, 075301 (2013)

Note that the damping of the spin is a purely dipolar beyond mean-field effect for a homogeneous system, associated with the growth of entanglement

> See also J. Ye (KRb molecules) Weidemüller (Rydberg atoms)

Spin-length data with and without lattice

Collective spin length $\sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2 + \langle S_z \rangle^2}$ from Ramsey interferometer

Lattice case : decrease of spin length due to dipolar interactions **quantum** thermalization

PRL 125, 143401 (2020)

Bulk case : Spins remain almost locked despite magnetic field gradient

preservation of ferromagnetism

Spin gap $\propto \frac{(a_6 - a_4)\hbar^2 n}{m}$

Classical ferrofluid

PRL 121, 013201 (2018)

Partial conclusions on the collective spin measurements

Strong decay of collective spin, associated with dipole-dipole interactions

The decays is « too » slow.

 \rightarrow **heating** in the lattice ?

 \rightarrow Are there more **holes** than we thought ?

 \rightarrow effect of **losses** ?

 \rightarrow more subtle effect associated with possibly disorder ?

(see glassy dynamics observed with Rydberg atoms Phys. Rev. X 11, 011011 (2021))

The measurement of coherences (the contrast of the interferometer) gives access to information we could not reach by simply measuring populations.

NB:
$$\sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2 + \langle S_z \rangle^2} \Big|_{t \to \infty} \approx 0$$
 can be related to $P_{ms} = \frac{1}{7} (1 + \beta B_Q (4 - m_s^2)) \approx exp[-\beta B_Q m_s^2]$

At equilibrium, the strongly interacting many-body system looks like a non-interacting one !

Outline:

1- Thermalization of the Zeeman populations

2- Thermalization of the collective spin

3- Experimental measurement of correlations using collective measurements

example: squeezing (see Ueda 1994)

What we learn from variances (ONLY for s>1/2)

$$\left\langle \hat{S}_{z}^{2} \right\rangle = \left\langle \left(\sum_{i=1}^{N} \hat{s}_{z,i} \right)^{2} \right\rangle = \left\langle \sum_{i=1}^{N} \hat{s}_{z,i}^{2} \right\rangle + \left\langle \sum_{i \neq j} \hat{s}_{z,i} \cdot \hat{s}_{z,j} \right\rangle$$

$$\text{VAR} \left(\hat{S}_{z} \right) + \sum_{i} \left\langle \hat{s}_{i}^{z} \right\rangle^{2} = \sum_{m_{s}} m_{s}^{2} P_{m_{s}} + \left\langle \sum_{i \neq j} \hat{s}_{z,i} \cdot \hat{s}_{z,j} \right\rangle - \sum_{i \neq j} \left\langle \hat{s}_{z,i} \right\rangle \cdot \left\langle \hat{s}_{z,j} \right\rangle$$

$$\text{Measured by the fluctuation of the collective spin (many images)}$$

$$Nm_{z}^{2} \text{ for a homogeneous system (many images)}$$

$$\text{CoVAR}(N_{m_{1}}N_{m_{2}}) = -NP_{m_{1}}P_{m_{2}} + \left\langle \sum_{i \neq j} \hat{n}_{m_{1},i} \cdot \hat{n}_{m_{2},j} \right\rangle - \sum_{i \neq j} \left\langle \hat{n}_{m_{1},i} \right\rangle \cdot \left\langle \hat{n}_{m_{2},j} \right\rangle$$

Measuring quantum variance from fluorescence imaging

Measuring quantum variance from fluorescence imaging

 \vec{B}_{ext}

Quantum

projection noise

 $Var(S_z) = \frac{3}{2}N$

Preparation noise $Var(Sz) \propto N^2$

Fit noise
$$Var(Sz) \propto 1$$

Photon shot noise

$$\frac{Noise}{Signal} = \sqrt{\frac{1}{\beta N}} \qquad Var(Sz) \propto \frac{1}{\beta}N$$

Photon collection efficiency

Currently: Technical Noise $(\alpha N^2) \sim .5 SQN(\alpha N)$ (@10000 Atoms)
Measuring quantum variance from fluorescence imaging



Measurement of connected two-body correlations



Bi-partite measurements





PRELIMINARY

Observation of anisotropic correlations due to anisotropy of dipolar interactions



See also W. Bakr (NaRb molecules under microscope)

 $2 CoVar(S_A^z, S_B^z) = Var(S^z) - Var(S_A^z) - Var(S_B^z) < 0$ for « classical enough system » where $Var(S^z) = 0$

Comparaisons to simulations

$\begin{array}{c} 6 \\ 4 \\ 2 \\ 0 \\ -2 \\ -4 \\ 0 \\ 5 \\ 10 \\ 15 \\ 20 \\ 25 \\ 30 \\ t (ms) \end{array}$

Second order cumulant approach

Gaussian Ansatz for S=3 Holstein-Primakokk Bosons

(Tommaso Roscilde)

GDTWA

(Ana Maria Rey, Sean Muleady)



PRELIMINARY

Some comments:

- These measurements are well-suited to macroscopic systems (but beware of the preparation noise)
- We have found new ways to experimentally characterize various two-body correlations for s>1/2
- Some of those are rather immune to technical/detection noise.
- But require lots of data
- We could maybe go to higher-order correlations using higher order moments (more difficult)

A list of over simplifications I've made:

- neglecting (dipolar relaxation) losses (can modify correlations)
- physics beyond singly-occupied sites rather unexplored.
- assuming inhomogeneity can be bad (here it looks ok)

and some questions:

- could we turn the measurements based on co-variances into entanglement witnesses?

See Irénée Frérot et al., arXiv:2203.13547 (2022)

Measuring objects smaller than resolution limit

-1

п_{1D}(у) (10² µm⁻⁷ т

0.2

1/7

0

-3

-2

 $\int_{0.1}^{s_{max}} d$

0.08 0.06 0.02O density (arb. units) 10 20 **CONCLUSION** +1/2+5/2-7/2 -3/2 -9/2 -4ħk -2ħk 2ħk 4ħk 0ħk -150 -50 50 150 Momentum y (μm) PRA 104, 033309 (2021) PRA 102, 013317 (2020) Quantum thermalization Measuring correlations Quantum thermalization of populations from (co-)variances of collective spin (b) 3.0 🛧 2.5 2.5 2.0 2.0 \mathcal{C} 1.5 1.5 1.0 Analytical formula 1.0 0.5 GDTWA 0.0 0.5 Sum ms^2 pms -1 0 2 3 0 20 40 60 80 100 experimental variance 0.0 m_S time (ms) 20 0 40 60 80 100 PRL 125, 143401 (2020) Nature Comm. 10, 1714 (2019)

PRL 121, 013201 (2018)

PRL 129, 023401 (2022)

A new spin-orbit scheme to

characterize SU(N) Fermi gases

Dipolar physics: A review of experiments with magnetic quantum gases arXiv:2201.02672









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Warm-up....

Measuring objects smaller than the imaging resolution

Measuring objects smaller than the imaging resolution





+ add diffraction



Idea: counting missing photons

$$Log\left(\frac{I}{I_0}\right) = -\int dz \, n(x, y, z)\sigma \coloneqq -OD(x, y)$$

X

y

$$OD(x,y) = OD_m(y)e^{-x^2/w^2}$$
 Ansatz

$$\frac{P(y) - P_0}{P_0} = \int dx \left(1 - e^{-OD_m(y)e^{-x^2/w^2}} \right)$$
$$= w \int du \left(1 - e^{-OD_m(y)e^{-u^2}} \right)$$

Deduce $OD_m(y)$ from experimentally measured missing photon number.

Need additional information

Add the requirement that $\int dx \, dy \, OD(x, y) = N \sigma$

Find w, and OD(y)



Short term dynamics of the many-body system



Perturbation theory

CONTRIBUTIONS TO FLUCTUATIONS



Data analysis: measure collective spins from probability distributions

$$\ell = \sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2 + \langle S_z \rangle^2}$$





Assume $\ell=0$, and Gaussian noise:





Method to derive ℓ : fit probability distributions with a convolution of the two distributions



Measuring the variance of magnetization



How many images?

 $m_s = 3$ $m_{s} = -3$ $m_s = 0$

N atoms provide N measurements for individual populations

N atoms provide 1 measurement for the collective spin S_{7}

$$P(x) = \frac{1}{\sqrt{2 \pi \sigma}} \exp\left(-\frac{(x - x_0)^2}{2\sigma^2}\right)$$

 $< x^2 > - < x > 2 = 2$

One image

 $< x^{4} > - < x^{2} > 2 = 2(\sigma^{4} + 2\sigma^{2}x_{0}^{2}))$

Standard deviation on the measurement of x^2 :

 $\sqrt{2(\sigma^4+2\sigma^2x_0^2)}$

 $x_0 \ll \sigma$ $\sqrt{\frac{2}{M}} \ll 1$

 $\frac{2x_0}{\sigma} \sqrt{\frac{1}{M}} \ll 1$ $x_0 \gg \sigma$ (M images)



Variance of the variance



$$Var(Mz) = Var_{exp} - Var_{rf} - Var_{fit} - Var_{shotnoise}$$

Histogram of the estimated variances Mean = 1.3 sdv of measurement = .86 Number of measurement = 28

 \rightarrow error of estimate .86/ $\sqrt{28}$ = .16

Asymptotic behavior



Take into account energy constraints.

Two contributions for energy

Dipole-dipole interactions

 $\langle \Psi(t)|V_{dd}|\Psi(t)\rangle$

Difficult to calculate except at t=0 Tensor light-shift leads to an effective quadratic Zeeman effect



 \rightarrow This explains why simply maximizing entropy is sufficient at small angles

Some results:

 Comparison between measured spin correlations and expected correlations for independent spins (red dots)



Idea 1: compare variance of Mz and Sum ms² Pms Interest: provides a correlation witness and a measurement of the correlations Weakness: sensitive to detection noise and rf noise

One measurement = Repeat 40 times the same experiment and measure Mz each time





Atoms are composite objects, whose spin can be larger than 1/2 $\vec{F} = \vec{S} + \vec{I}$



From 2 to N atoms



In the mean-field approximation, atoms undergo (classical) precession Mean-field may be inhomogeneous, and total spin may not be conserved

With and without lattice: the main difference (Lanczos approach)

Lattice case :

$$H_{dd} = -\frac{1}{2}S'_{x} \cdot S'_{x} - \frac{3}{8}(S'^{+}S'^{+} + S'^{-}S'^{-}) + \frac{1}{8}[(S'^{+}S'^{-} + S'^{-}S'^{+})]$$

$$\Psi_{0} = |3_{x}, 3_{x}, 3_{x}, 3_{x}, \dots, 3_{x}\rangle \xrightarrow{T} \qquad (...)$$
BEC case :

$$\Psi_{0} = |3_{x}, 3_{x}, 3_{x}, 3_{x}, \dots, 3_{x}\rangle \xrightarrow{T} \qquad \Psi_{1} = (2_{x}, 2_{x}) \cdot 3_{x}, 3_{x}, \dots, 3_{x}, 3_{x}, 3_{x}, \dots, 3_{x}) \xrightarrow{T} \qquad (...)$$
Spin gap $\propto \frac{(a_{6} - a_{4})\hbar^{2}n}{m}$

In the BEC case, protection of ferromagnetism after the quench due to a spin gap

Quench results in the excitation of trapped of magnon mode (and the retardation of thermalization)



Outlook : Quantum thermalization, entanglement

Reduced density matrix (isolate one spin and trace over the rest of the system)



(calculations by J. Schachenmayer)

Tr(ho^2)=1

 $\operatorname{Tr}((\rho|_A)^2) \neq 1$

What we learn from co-variances (S>1/2 only)

$$CoVAR(N_{m_1}N_{m_2}) = \left\langle \hat{N}_{m_1}\hat{N}_{m_2} \right\rangle - \left\langle \hat{N}_{m_1} \right\rangle \left\langle \hat{N}_{m_2} \right\rangle$$

$$\left\langle \hat{N}_{m_1}\hat{N}_{m_2} \right\rangle = \left\langle \sum_{i=1}^{N} \hat{n}_{m_1,i}.\hat{n}_{m_2,i} \right\rangle + \left\langle \sum_{i \neq j} \hat{n}_{m_1,i}.\hat{n}_{m_2,j} \right\rangle$$

$$0 \text{ for single sites}$$

$$CoVAR(N_{m_1}N_{m_2}) = -NP_{m_1}P_{m_2} + \left\langle \left\langle \sum_{i \neq j} \hat{n}_{m_1,i}.\hat{n}_{m_2,j} \right\rangle - \sum_{i \neq j} \left\langle \hat{n}_{m_1,i} \right\rangle . \left\langle \hat{n}_{m_2,j} \right\rangle$$

$$Assumption: homogeneous populations$$

$$\sum_i \left\langle \hat{n}_{m_1,i} \right\rangle . \left\langle \hat{n}_{m_2,i} \right\rangle = NP_{m_1}P_{m_2}$$

Cold atoms revisit condensed matter physics

Optical lattices

Perriodic potential introduced by stationnary wave





Atoms in optical lattices can mimick electrons in solids

Quantum Magnetism

Fermionic isotope in the ground state: SU(N) symmetry



Spin entirely due to nucleus

Spin-independent interactions

One consequence : no spin-exchange dynamics

- Can prepare arbitrary number of (fixed) « colours »

Proposal : interplay between SU(N) magnetism and lattice topology



3 colors Triangular lattice Ordered

2 colors

Square Ordered



Frederic Mila

For a square lattice: SU(2) ordered SU(3 and 4) disordered SU(5) ordered (very low T's) SU(6) disordered... Honeycomb and Kagomé lattice very interesting for SU(N=3,4).

SU(2) : 2 atom singlet SU(4) : 4-atom singlet (see Takahashi)

NB: correlations arrise at higher entropy!!

Partial conclusions on the collective spin measurements

Strong decay of collective spin, associated with dipole-dipole interactions

The decays is « too » slow.

 \rightarrow **heating** in the lattice ?

 \rightarrow Are there more **holes** than we thought ?

 \rightarrow effect of **losses** ?

 \rightarrow more subtle effect associated with possibly disorder ?

(see glassy dynamics observed with Rydberg atoms Phys. Rev. X 11, 011011 (2021))

The measurement of coherences (the contrast of the interferometer) gives access to information we could not reach by simply measuring populations.

NB:
$$\sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2 + \langle S_z \rangle^2} \Big|_{t \to \infty} \approx 0$$
 can be related to $P_{ms} = \frac{1}{7} (1 + \beta B_Q (4 - m_s^2)) \approx exp[-\beta B_Q m_s^2]$

At equilibrium, the strongly interacting many-body system looks like a non-interacting one !

Damping of the collective spin due to dipolar interactions



Good agreement at short times Good agreement with second-order perturbation theory too

At long times, the collective spin decays **SLOWER** than it should !!

Two realizations with cold atoms

Ground state atoms in optical lattices and super-exchange interactions



Rydberg atoms in tweezers and dipole-dipole interactions

$$\alpha S_{1z} S_{2z} + \beta \frac{1}{2} \left(S_{1+} S_{2-} + S_{1-} S_{2+} \right)$$





M. Greiner's experiment (see also Hulet, Kohl, Zwierlein, Bloch...)

(Browaeys, ...)

Two realizations with cold atoms

Ground state atoms in optical lattices and super-exchange interactions



Atoms or molecules with dipole-dipole interactions





 $-\frac{1}{4}(S_{1+}S_{2-}+S_{1-}S_{2+})$

Ising

Exchange

How to characterize a quantum many-body system

Partial information : trace over subsystem

Example for a pure state:

 $\rho = |\Psi\rangle\langle\Psi|$

« pure »

 $Tr(\rho^2)=1$

 $\operatorname{Tr}((\rho|_{A})^{2})\neq 1$

« locally mixed »

Measures the entropy associated to entanglement

See Greiner 2016 Klempt; Treutlein

$$\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle+|\downarrow\uparrow\rangle)$$

$$\left(\frac{1}{\sqrt{2}}(|\uparrow\rangle+|\downarrow\rangle)\right)^2$$

Basic idea $\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$

But measurement performed in just one lattice site will show random fluctuations $\langle m_s = \pm 1/2 \rangle \rightarrow$ associated entropy
Length of spin vs correlations – Two Atoms





Two initially polarized atoms (A and B):



 $\vec{B}_A(B) = \vec{B}_B(A) +$

Classically:

Quantum-mechanically:

These two atoms undergo identical precession

Total spin is conserved in time

Possibility for entanglement

Total spin is NOT conserved

Measuring interatomic correlations through covariances

$$CoVAR(N_{m_1}N_{m_2}) = -NP_{m_1}P_{m_2} + \left\langle \sum_{i \neq j} \hat{n}_{m_1,i} \cdot \hat{n}_{m_2,j} \right\rangle - \sum_{i \neq j} \left\langle \hat{n}_{m_1,i} \right\rangle \cdot \left\langle \hat{n}_{m_2,j} \right\rangle$$

Measured by
fluctuations
(many images) Assumption: homogeneous populations
$$\sum_{i} \left\langle \hat{n}_{m_1,i} \right\rangle \cdot \left\langle \hat{n}_{m_2,i} \right\rangle = NP_{m_1}P_{m_2}$$

A few general properties – correlations in the steady « thermalized » state

NB:
$$P_{m_s} = \frac{1}{7} \left(1 + \beta B_Q (4 - m_s^2) \right) \qquad \text{for simplicity we focus on } B_Q = 0 \rightarrow P_{m_s} = 1/7 \qquad \sum m_s^2 P_{m_s} = \frac{9}{2} N$$

 $[S_z, H] = 0$

$$\Psi_{ini} = (|ms = 3\rangle_x)^N \qquad \qquad VAR(S_z) = \frac{3}{2}N \qquad \qquad \sum_{i \neq j} \langle S_i^z S_j^z \rangle - \langle S_i^z \rangle \langle S_j^z \rangle = -\frac{5}{2}N$$

$$\Psi_{ini} = (1/\sqrt{2}(|ms = 3\rangle + |ms = -3\rangle))^N \qquad \qquad VAR(S_z) = 9N \qquad \qquad \sum_{i \neq j} \langle S_i^z S_j^z \rangle - \langle S_i^z \rangle \langle S_j^z \rangle = \frac{9}{2}N$$

S=1/2 or mixtures :
$$\sum_{i \neq j} \langle S_i^z S_j^z \rangle - \langle S_i^z \rangle \langle S_j^z \rangle = 0$$