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Supplementary Material for

Entanglement between two spatially separated atomic modes

Karsten Lange, Jan Peise, Bernd Lücke, Ilka Kruse, Giuseppe Vitagliano, Iagoba Apellaniz, Matthias Kleinmann, Géza Tóth, Carsten Klempt*

*Corresponding author. E-mail: klempt@iqo.uni-hannover.de

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Supplementary Text Fig. S1 References

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Experimental sequence and analysis. The experimental procedure and evaluation, as well as a discussion of the number-dependent detection noise can be found in detail in the supplemental material of Ref. (*16*).

We alternate between the experiments for the two measurement directions J_z and J_{\perp} to minimize the influence of changing ambiance conditions. Both measurements start with the same experimental sequence. A BEC is prepared in a crossed-beam optical dipole trap in the state $F = 1, m_F = 0$.

A red-detuned microwave dressing field with a detuning of 206 kHz couples the levels F = $1, m_F = -1$ and $F = 2, m_F = -2$. This induces an energy shift of the levels such that a resonance condition for spin-changing collisions from $F = 1, m_F = 0$ to $F = 1, m_F = \pm 1$ is reached (16). At the resonance, the energy of two atoms in the $m_F = 0$ state is equal to the energy of two atoms in $m_F = \pm 1$ plus the energy of the excitation to the first spatially excited mode. This pair creation process, producing a pair of entangled atoms in $m_F = \pm 1$, is subject to bosonic enhancement, creating further pairs in the same mode during an interaction time of t = 180 ms. Due to the nature of the spin-changing collisions, the $F = 1, m_F = \pm 1$ levels are populated with a two-mode squeezed state. The two-mode squeezed state consists of a superposition of twin-Fock states $\sum_n c_n |n\rangle_{+1} |n\rangle_{-1}$ with an equal number of atoms $N_{\pm 1}$ in the two Zeeman levels $m_F = \pm 1$. The weight $c_n = \frac{(-i \tanh \xi)^n}{\cosh \xi}$ corresponds to a squeezing strength $\xi = \Omega t$ and a spin dynamics rate $\Omega = 2\pi \times 6.6$ Hz. The final measurement of the total number of atoms collapses the state onto a twin-Fock state. The measurement of J_z is now a measurement of the atom numbers in the two levels $m_F = \pm 1$ of the F = 1 manifold. However, to keep the two experimental procedures as similar as possible, the ensembles are transferred to the F = 2 manifold before detection. To this end, the pulse sequence of the transfer pulses (II-IV) is reversed for the J_z measurements with respect to the J_{\perp} measurement (see Fig. S1).

The measurement in the orthogonal direction requires a rotation around an axis perpendicular to J_z . This is achieved by a coupling of the ensembles in F = 1, $m_F = \pm 1$ by an effective $\pi/2$ pulse. Since the microwave phase is not synchronized to the atomic phases, the rotation leads to a measurement of J_{\perp} along an arbitrary direction in the x - y plane. However, the state is fully characterized due to its perfect symmetry under rotations around the z axis. Firstly, an ideal twin-Fock state is symmetric itself, and secondly, the experimentally realized state is randomized over J_z rotations due to the influence of magnetic field noise. Therefore, the measurement of J_{\perp} is sufficient. The detection is again realized in the F = 2 manifold with the $m_F = \pm 1$ ensembles occupying F = 2, $m_F = -2$ and F = 2, $m_F = 0$. The large condensate from $m_F = 0$ is mainly transferred to F = 2, $m_F = -1$ with a small fraction transferred to F = 2, $m_F = 1$.

The experimental sequence ends with the detection of the atomic ensembles. The dipole trap is switched off to allow for 7.5 ms of self-similar expansion. The mode profiles remain undistorted but magnified due to the interaction of the ensembles with the large condensate remaining in the F = 2, $m_F = -1$ state (40). After the initial mean-field dominated expansion, a strong magnetic field gradient is applied to spatially separate the atoms in the populated Zeeman levels. Finally, the number of atoms in the clouds is detected by absorption imaging on a CCD camera with a large quantum efficiency.

The absorption images are used to detect the number of atoms in the two spatially separated clouds. The center of mass of the large condensate in the F = 2, $m_F = -1$ level is used as a reference for the position of all clouds (see Fig. S1B). This is necessary due to slight shot-to-shot variations of the position, which result from minute position changes of the dipole trap. The position of the masks for the ensembles in F = 2, $m_F = \{-2, 0\}$ (formerly F = 1, $m_F = \pm 1$), as well as the cutting line for the parts a (left) and b (right), is fixed with respect to this reference.

The number of atoms in the four resulting sub-masks is then obtained by summing over the column density of the absorption image. The detection noise amounts on average to 24 atoms. It is independent of the particular number and is obtained by simultaneously evaluating empty images. The squeezed variance $(\Delta J_z)^2$ is calculated by subtracting the detection noise variance from the raw atomic variance.

To utilize the created state for quantum information tasks, it can be transferred into an optical lattice, where all constituent particles are individually addressable. As a concrete example, single-atom projective measurements on one half of this highly entangled ensemble allow to synthesize any pure symmetric quantum state in the second half (*35,36*).

Bootstrapping. The error bars in Figs. 3 and 4 are obtained via a bootstrapping method. We created 10,000 random data sets on the basis of the distributions of the experimental data. We then calculated the standard deviations of the measured quantities from these 10,000 samples and checked that the percentage of violations of equation (1) was consistent with the reported significance.

Proof of equation (1). We start from the sum of two Heisenberg uncertainty relations $(\Delta J_z)^2[(\Delta J_x)^2 + (\Delta J_y)^2] \ge \frac{1}{4}(\langle J_x \rangle^2 + \langle J_y \rangle^2)$. Simple algebra yields

$$\left[(\Delta J_{z})^{2} + \frac{1}{4} \right] \times \frac{(\Delta J_{x})^{2} + (\Delta J_{y})^{2}}{\langle J_{x}^{2} \rangle + \langle J_{y}^{2} \rangle} \ge \frac{1}{4}.$$
(S1)

Here, the first factor represents the fluctuations in the particle number difference and the second term represents the fluctuations in the phase difference.

Product states. First, we consider product states of the form $|\Psi^{(a)}\rangle \otimes |\Psi^{(b)}\rangle$. For such states

$$\begin{bmatrix} (\Delta J_{z}^{+})^{2} + \frac{1}{2} \end{bmatrix} \times \begin{bmatrix} (\Delta \tilde{J}_{x}^{-})^{2} + (\Delta \tilde{J}_{y}^{-})^{2} \end{bmatrix}$$

= $[(\mathcal{U}^{(a)} + \frac{1}{4}) + (\mathcal{U}^{(b)} + \frac{1}{4})] \cdot (\mathcal{V}^{(a)} + \mathcal{V}^{(b)})$
$$\geq 4\sqrt{(\mathcal{U}^{(a)} + \frac{1}{4})(\mathcal{U}^{(b)} + \frac{1}{4})\mathcal{V}^{(a)}\mathcal{V}^{(b)}} \geq 1$$
 (S2)

holds, where we used the notation $\mathcal{U}^{(n)} = (\Delta J_z^{(n)})^2$ and $\mathcal{V}^{(n)} = (\Delta \tilde{J}_x^{(n)})^2 + (\Delta \tilde{J}_y^{(n)})^2$ for n = a, b. For product states, the variance of a collective observable is the sum of the subsystem variances, i.e. $[\Delta (A^{(a)} + A^{(b)})]^2 = (\Delta A^{(a)})^2 + (\Delta A^{(b)})^2$, leading to the equality in equation (S2). The first inequality is obtained from the inequality between the arithmetic and the geometric mean. Equation (S1) is valid for both part a and b of the state, leading to the second inequality.

Using $\langle (\tilde{J}_{x}^{(n)})^{2} \rangle + \langle (\tilde{J}_{y}^{(n)})^{2} \rangle = 1$ for n = a, b, equation (S2) yields

$$2\left[(\Delta J_{z}^{+})^{2} + \frac{1}{2}\right](\mathcal{S} - \mathcal{C}) \ge \mathcal{S},$$
(S3)

where correlations between the two subsystems are characterized by $C = \left\langle \frac{J_x^{(a)} J_x^{(b)} + J_y^{(a)} J_y^{(b)}}{j_a j_b} \right\rangle$, and $S = \mathcal{J}^{(a)} \mathcal{J}^{(b)}$. Note that C can be negative and $|C| \leq S$. The normalization with the total spin will make it easier to adapt our criterion to experiments with a varying particle number in the ensembles.

Separable states. We now consider a mixed separable state of the form $\rho_{sep} = \sum_k p_k |\Psi_k^{(a)}\rangle \otimes |\Psi_k^{(b)}\rangle$. For such states, we can write the following series of inequalities

$$2\left[(\Delta J_{z}^{+})^{2} + \frac{1}{2} \right] (S - C)$$

$$\geq 2\left[\sum_{k} p_{k} (\Delta J_{z})_{k}^{2} + \frac{1}{2} \right] \left[\sum_{k} p_{k} (S_{k} - C_{k}) \right]$$

$$\geq 2\left[\sum_{k} p_{k} \sqrt{\left((\Delta J_{z})_{k}^{2} + \frac{1}{2} \right) (S_{k} - C_{k})} \right]^{2}$$

$$\geq \left(\sum_{k} p_{k} \sqrt{S_{k}} \right)^{2}, \qquad (S4)$$

where the subscript k indicates that the quantity is computed for the k^{th} sub-ensemble $|\Psi_k^{(a)}\rangle \otimes$ $|\Psi_k^{(b)}\rangle$. The first inequality in equation (S4) is due to $(\Delta J_z^+)^2$ and S being concave in the quantum state. The second inequality is based on the Cauchy-Schwarz inequality $(\sum_k p_k a_k)$ $(\sum_k p_k b_k) \ge$ $\left(\sum_{k} p_k \sqrt{a_k b_k}\right)^2$, where $a_k, b_k \ge 0$. The third inequality is the application of equation (S3) for all sub-ensembles. Next, we find a lower bound on the RHS of equation (S4) based on the knowledge of $\mathcal{J}^{(a)}$ and $\mathcal{J}^{(b)}$. We find that

$$\sum_{k} p_k \left(\mathcal{J}_k^{(a)} \mathcal{J}_k^{(b)} \right)^{1/2} \ge (\mathcal{J}^{(a)})^2 + (\mathcal{J}^{(b)})^2 - 1,$$
(S5)

which is based on noting $(xy)^{1/4} \ge x + y - 1$ for $0 \le x, y \le 1$. Using equation (S5) to bound the RHS of equation (S4) from below and dividing by S we obtain

$$\left[(\Delta J_{z}^{+})^{2} + \frac{1}{2} \right] \times \left[2 - 2\frac{\mathcal{C}}{\mathcal{S}} \right] \geq \frac{\left[(\mathcal{J}^{(a)})^{2} + (\mathcal{J}^{(b)})^{2} - 1 \right]^{2}}{\mathcal{S}}.$$
(S6)

Non-zero particle number variance. So far, we assumed that the particle number of the two clouds a and b are known constants. In practice, the particle number is not a constant, but varies from experiment to experiment. In principle, one could postselect experiments for a given particle number, and test entanglement only in the selected experiments. However, this leads to discarding most experiments, increasing the number of repetitions needed tremendously. Hence, we modify our condition to handle non-zero particle number variances (41). In this case, the state of the system can be written as $\rho = \sum_{j_a,j_b} Q_{j_a,j_b} \rho_{j_a,j_b}$, where ρ_{j_a,j_b} are states with $2j_a$ and $2j_b$ particles in the two clouds, $Q_{j_a,j_b} \ge 0$, $\sum_{j_a,j_b} Q_{j_a,j_b} = 1$. The state ρ is separable if and only if all ρ_N are separable. Then, expectation values for ρ are computed as $\langle Af(\hat{j}_a, \hat{j}_b) \rangle_{\rho} = \sum_{j_a,j_b} Q_{j_a,j_b} \langle A \rangle_{\rho_{j_a,j_b}} f(j_a, j_b)$, where the operator is separated into one part that depends only on the particle number operators of the two clouds represented by \hat{j}_a and \hat{j}_b , and another part that does not depend on them. f(x) denotes some function. The proof from equation (S2) to equation (S6) can then be repeated, assuming that j_a and j_b are operators.

Note that we choose the normalization of the variances such that the criterion is robust against fluctuations of the total number of particles. For a constant particle number one could simplify the fractions on the LHS of equation (1) by multiplying both the denominator and the numerator by $j_{\rm a}$, and for the other fractions by $j_{\rm b}$.

Supplementary figures



Figure S1: **Experimental techniques.** (A) Experimental procedure for the measurement of J_{\perp} . The sequence starts with a BEC in the $|F = 1, m_F = 0\rangle$ Zeeman level. (I) A microwave dressing field (solid blue line) induces a resonance condition for spin-changing collisions, allowing for the population of the $|1, \pm 1\rangle$ levels with a twin-Fock state (dashed blue line). (II) The population in $|1, 1\rangle$ is completely transferred to $|2, 0\rangle$ via a microwave transfer (solid orange arrows). This also leads to a population of $|2, 1\rangle$ with atoms from $|1, 0\rangle$ (dotted orange line). (III) A coupling between the two levels populated by the twin-Fock state is induced by a $\pi/2$ microwave pulse (solid dark blue line). This coupling is also resonant to the transition $|1, 0\rangle \rightarrow |2, -1\rangle$, populating the $|2, -1\rangle$ level (dotted dark blue line). (IV) To avoid an overlap in the detection of the atoms in $|1, -1\rangle$ and $|2, 1\rangle$ the ensemble from $|1, -1\rangle$ is transferred to $|2, -2\rangle$ (solid dark orange line). (B) Single-shot absorption image with read-out masks indicated in orange. The position of the masks is determined by the center of mass of the central atomic cloud. The atom numbers in all four sub-masks are evaluated by summing over the column densities in the appropriate sub-masks.

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