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Quantum Atom Optics with Fermions from Molecular Dissociation

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We study a fermionic atom optics counterpart of parametric down-conversion with photons. This can be realized through dissociation of a Bose-Einstein condensate of molecular dimers consisting of fermionic atoms. We present a theoretical model describing the quantum dynamics of dissociation and find analytic solutions for mode occupancies and atomic pair correlations, valid in the short time limit. The solutions are used to identify upper bounds for the correlation functions, which are applicable to any fermionic system and correspond to ideal particle number-difference squeezing.

Advances in the experimental control of degenerate quantum gases of neutral atoms have recently reached the stage where atomic correlations and quantum statistics can be *directly* accessed via the measurement of atom shot noise and atom counting [1–5]. Given the similarities with the pioneering photon correlation measurements of Hanbury Brown and Twiss [6] and the intriguing parallels with modern quantum optics, these experiments represent a remarkable step forward in advancing quantum atom optics. Earlier and related experiments were performed either in a cold but not degenerate atomic beam [7] or else were examples of *indirect* measurements of higher-order correlations and number squeezing [8–11].

In quantum optics with photons the most successful applications have been achieved using squeezed light and entangled photon pairs from parametric down-conversion [12–15]. A matter wave or atom optics counterpart of down-conversion can be realized through dissociation of a Bose-Einstein condensate (BEC) of molecular dimers. The most direct analogy with quantum optics corresponds to the case of bosonic statistics of constituent atom pairs [16–19], as realized in dissociation experiments with ^{23}Na$_2$ and ^{87}Rb$_2$ [20,21]. In contrast to this, the recent correlation measurements at JILA [1], using dissociation of ^{40}K$_2$ molecules near a magnetic Feshbach resonance, have an intriguing twist in that the constituent atoms obey fermionic statistics. In this case, analogies with photonic down-conversion and the implications for possible future applications are not so immediate as in the case of bosons.

In this Letter we study the quantum dynamics of dissociation of diatomic molecules into fermionic atoms and analyze the resulting atom correlations in momentum space. At low densities and small *s*-wave interactions the momentum correlations are reflected in the density correlations after spatial expansion of the cloud. Accordingly, the results obtained here are related to the spatial correlation measurements performed at JILA [1]. Together with the recent studies of fermionic four-wave mixing [22] and association of fermionic atoms into molecules [23,24], the present work (see also [25]) expands the paradigm of

fermionic quantum atom optics. Possible applications are in precision measurements and fundamental tests of quantum mechanics, similar to those proposed recently for dissociation into bosonic atoms [17,19].

Because of the limitations of the undepleted molecular field approximation employed here, the obtained results are only applicable to short dissociation times. However, the advantage is the analytic transparency of the results, which provide useful insights at the conceptual level. In particular, we show rigorously that the obtained pair correlations represent their generic upper bounds and are applicable to any fermion system. This gives a useful reference for further (numerical) studies of this and related systems with less restricted approximations. In addition, we point out that the notions of maximum correlation and squeezing of atom number-difference fluctuations have to take into account the fact that the shot-noise level for fermions is fundamentally different to what one usually encounters in quantum optics with bosons.

We start the analysis by considering an effective field theory Hamiltonian for the coupled atomic-molecular system given by $\hat{H} = \hat{H}_0 - i\hbar\chi \int d\mathbf{x}(\hat{\Psi}_0^\dagger \hat{\Psi}_\uparrow \hat{\Psi}_\downarrow - \hat{\Psi}_\downarrow^\dagger \hat{\Psi}_\uparrow^\dagger \hat{\Psi}_0)$ [26]. Here, $\hat{H}_0$ stands for the usual kinetic energy term plus the trapping potential, $\hat{\Psi}_0(\mathbf{x}, t)$ is the bosonic field operator for molecules of mass m_0, while $\hat{\Psi}_{\uparrow(\downarrow)}(\mathbf{x}, t)$ are fermionic operators for atoms (with masses $m_{\uparrow(\downarrow)} = m_0/2 \equiv m$) in two different spin states, $\sigma = \uparrow, \downarrow$. The atom-molecule coupling is described by χ, and we have omitted intra- and interspecies *s*-wave scattering interaction terms, which is justified at low particle densities.

Considering a uniform system in a cubic box of side *L*, we employ an undepleted molecular field approximation valid in the short time limit. The molecular field is described by a coherent state and we absorb its mean-field amplitude Ψ_0 into an effective coupling $g = \chi\sqrt{n_0}$, where $n_0 = |\Psi_0|^2$ is the density. Assuming periodic boundary conditions and expanding the atomic fields in a plane-wave basis, in terms of single-mode operators, we obtain the following effective Hamiltonian, in a rotating frame:

$$\hat{H} = \sum_{\mathbf{k},\sigma} \hbar\Delta_{\mathbf{k}} \hat{n}_{\mathbf{k}\sigma} - i\hbar g \sum_{\mathbf{k}} (\hat{c}_{\mathbf{k}\uparrow}\hat{c}_{-\mathbf{k}\downarrow} - \hat{c}^{\dagger}_{-\mathbf{k}\downarrow}\hat{c}^{\dagger}_{\mathbf{k}\uparrow}). \quad (1)$$

Here, $\hat{c}^{\dagger}_{\mathbf{k}\sigma}$ ($\hat{c}_{\mathbf{k}\sigma}$) are fermionic creation (annihilation) operators, $\hat{n}_{\mathbf{k}\sigma} = \hat{c}^{\dagger}_{\mathbf{k}\sigma}\hat{c}_{\mathbf{k}\sigma}$ is the particle number operator, $\mathbf{k}$ is the momentum [$k_i = 2\pi n_i/L$, $n_i = 0, \pm 1, \pm 2, \dots$, $i = x, y, z$], and $\Delta_{\mathbf{k}} \equiv \Delta + \hbar k^2/(2m)$. The detuning 2Δ corresponds to the overall energy mismatch $2\hbar\Delta$ between the free two-atom state in the dissociation threshold and the bound molecular state.

The model system in mind corresponds to a pure molecular BEC on the stable side of a Feshbach resonance, with no residual atoms present. This is followed by a rapid switching on of the coupling χ (e.g., via an rf transition or a rapid crossing through the resonance to the atomic side) and a simultaneous switching off of the trapping potential. From this stage onward, the atomic filed evolves in free space, with a vacuum initial state and negative detuning Δ. Since the excess of energy $2\hbar|\Delta|$ is released into the kinetic energy of dissociated atom pairs in the two spin states, $2\hbar|\Delta| \to \hbar^2(|\mathbf{k}_{\uparrow}|^2 + |\mathbf{k}_{\downarrow}|^2)/(2m)$, we expect—from momentum conservation—strong correlation between the atoms with opposite spins and momenta, $\mathbf{k}_{\uparrow} = -\mathbf{k}_{\downarrow}$. In fact, the interaction term in the Hamiltonian (1) is the prototype interaction to produce—in the lowest order perturbation theory—an entangled spin singlet state. It is also a fermionic analog of the squeezing Hamiltonian in quantum optics [15].

Introducing a dimensionless time $\tau = t/t_0$, length $l = L/d_0$, detuning $\delta = \Delta t_0$, and momentum $\mathbf{q} = \mathbf{k}d_0$, where $t_0 = 1/g$ is the time scale and $d_0 = \sqrt{\hbar t_0/(2m)}$ is the length scale, we can put the system into a dimensionless form, with the Heisenberg equations of motion:

$$d\hat{c}_{\mathbf{q}\uparrow}/d\tau = -i\delta_{\mathbf{q}}\hat{c}_{\mathbf{q}\uparrow} - \hat{c}^{\dagger}_{-\mathbf{q}\downarrow}, \quad (2a)$$

$$d\hat{c}^{\dagger}_{-\mathbf{q}\downarrow}/d\tau = i\delta_{\mathbf{q}}\hat{c}^{\dagger}_{-\mathbf{q}\downarrow} + \hat{c}_{\mathbf{q}\uparrow}, \quad (2b)$$

where $\delta_{\mathbf{q}} \equiv q^2 + \delta$ ($q = |\mathbf{q}|$). Solutions to Eq. (2) are

$$\hat{c}_{\mathbf{q}\uparrow}(\tau) = A_{\mathbf{q}}(\tau)\hat{c}_{\mathbf{q}\uparrow}(0) - B_{\mathbf{q}}(\tau)\hat{c}^{\dagger}_{-\mathbf{q}\downarrow}(0), \quad (3a)$$

$$\hat{c}^{\dagger}_{-\mathbf{q}\downarrow}(\tau) = B_{\mathbf{q}}(\tau)\hat{c}_{\mathbf{q}\uparrow}(0) + A^{*}_{\mathbf{q}}(\tau)\hat{c}^{\dagger}_{-\mathbf{q}\downarrow}(0). \quad (3b)$$

Here, $A_{\mathbf{q}}(\tau) = \cos(g_{\mathbf{q}}\tau) - i\delta_{\mathbf{q}}\sin(g_{\mathbf{q}}\tau)/g_{\mathbf{q}}$, $B_{\mathbf{q}}(\tau) = \sin(g_{\mathbf{q}}\tau)/g_{\mathbf{q}}$, $g_{\mathbf{q}} \equiv (1 + \delta_{\mathbf{q}}^2)^{1/2}$, and $|A_{\mathbf{q}}|^2 + B_{\mathbf{q}}^2 = 1$. Using these solutions with vacuum initial conditions, we find that the only nonzero second-order moments are the mode occupancies and the pairing fields:

$$n_{\mathbf{q}}(\tau) \equiv \langle \hat{n}_{\mathbf{q}\sigma}(\tau) \rangle = B_{\mathbf{q}}^2(\tau) = \sin^2(g_{\mathbf{q}}\tau)/g_{\mathbf{q}}^2, \quad (4)$$

$$m_{\mathbf{q}}(\tau) \equiv \langle \hat{c}_{\mathbf{q}\uparrow}(\tau)\hat{c}_{-\mathbf{q}\downarrow}(\tau) \rangle = A_{\mathbf{q}}(\tau)B_{\mathbf{q}}(\tau), \quad (5)$$

which, in addition, are related by

$$|m_{\mathbf{q}}(\tau)|^2 = [1 - n_{\mathbf{q}}(\tau)]n_{\mathbf{q}}(\tau). \quad (6)$$

For comparison, in the case of bosons the last terms in Eqs. (2a) and (3a) acquire positive signs, while the sin

(cos) terms in the coefficients $A_{\mathbf{q}}$ and $B_{\mathbf{q}}$ are replaced by sinh (cosh), together with $g_{\mathbf{q}} \equiv (1 - \delta_{\mathbf{q}}^2)^{1/2}$ and $|A_{\mathbf{q}}|^2 - B_{\mathbf{q}}^2 = 1$ [27]. As a result, the solutions give $n_{\mathbf{q}}(\tau) = \sinh^2(g_{\mathbf{q}}\tau)/g_{\mathbf{q}}^2$ and $|m_{\mathbf{q}}(\tau)|^2 = [1 + n_{\mathbf{q}}(\tau)]n_{\mathbf{q}}(\tau)$.

From Eq. (4) we see that the momentum distribution of the atoms in the two spin states, $\uparrow$ and $\downarrow$, is the same. The average mode occupancies undergo oscillations characteristic of fermionic statistics [see Fig. 1(a)]; the maximum occupancy of $n_{\mathbf{q}}(\tau) = 1$ imposed by the Pauli exclusion principle is reached at integer multiples of time $\tau = \pi/2$, for resonant modes satisfying $g_{\mathbf{q}_0} = 1$. For $\mathbf{q}_0$ to be nonzero, this condition requires a negative detuning δ, and therefore the absolute resonant momentum is given by $q_0 = |\mathbf{q}_0| = \sqrt{|\delta|}$. During the initial stage ($\tau \lesssim 0.6$), the occupancies grow in phase and the 3D momentum distribution is peaked on the surface of a spherical shell of radius q_0 as shown in the inset of Fig. 1(a). At later times the oscillations dephase and the distribution function becomes more complicated in structure.

The total number of atoms in each spin state, $N_{\sigma}(\tau) = \sum_{\mathbf{q}} n_{\mathbf{q}}(\tau)$, as a function of time is shown in Fig. 1(b). The initial growth of $N_{\sigma}(\tau)$ saturates at $\tau \simeq 1.2$, after which we see nontrivial oscillations. This is a combined effect of Pauli blocking and the oscillatory behavior of the individual mode occupancies. We emphasize that the saturation in this model is obtained within the undepleted molecular field approximation, and thus is purely a consequence of Fermi statistics. By comparison, the same approximation for bosons leads to an exponentially growing output due to bosonic stimulation and hence to unphysical results in the long time limit. Here, once the depletion is taken into account, the saturation is naturally reached due to a finite initial number of molecules.
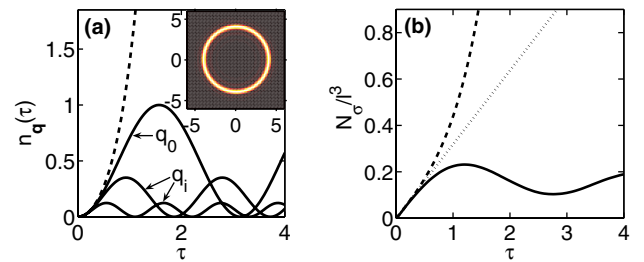

FIG. 1 (color online). (a) Average occupancies of one of the resonant modes $q_0 = \sqrt{|\delta|}$ and two sidebands q_i (solid lines) as a function of time, for $\delta = -16$. The dashed line is the solution for the q_0 mode in the case of bosons, which grows exponentially. The inset shows the slice through the origin of the fermionic 3D momentum distribution $n_{\mathbf{q}}(\tau)$ at $\tau = 0.6$. (b) Total number of atoms N_{σ}/l^3 (solid line) as a function of time, for $\delta = -16$. The normalization with respect to l^3 makes this quantity independent of the quantization volume. The dashed line is the respective bosonic result, while the straight dotted line is the total number of atoms $N_{\sigma}(\tau)/l^3 \simeq \sqrt{|\delta|}\tau/(4\pi)$ obtained using Fermi's golden rule calculation of the molecular spontaneous decay rate [20] in the linear regime.

In this respect, the present fermionic results suggest that the undepleted molecular field approximation for fermions is more reliable than for bosons. At the level of pairwise mode coupling, this conjecture is in fact supported by the results of a numerical simulation using an exact quantum Monte Carlo method [28]. For example, at $\tau \simeq \pi/2$ the discrepancy between the present and the exact results is about 5% for fermions, while it is ~11% for bosons and grows further with time. Similarly, the negligible role of the molecular depletion in the short time limit can be verified using the results of a related numerical study of Ref. [25] where the molecular field dynamics is treated at the mean-field level.

We now turn to the analysis of pair correlations of the atoms in the opposite spin states and consider a normalized correlation function between 3D density fluctuations $\Delta\hat{n}_{\mathbf{q}\sigma} = \hat{n}_{\mathbf{q}\sigma} - \langle\hat{n}_{\mathbf{q}\sigma}\rangle$ in momentum space:

$$g_{\uparrow\downarrow}(\mathbf{q}, \mathbf{q}', \tau) = \langle\Delta\hat{n}_{\mathbf{q}\uparrow}\Delta\hat{n}_{\mathbf{q}'\downarrow}\rangle / \sqrt{\langle\hat{n}_{\mathbf{q}\uparrow}\rangle\langle\hat{n}_{\mathbf{q}'\downarrow}\rangle}. \quad (7)$$

For atom pairs with nonopposite momenta the pair correlation vanishes, $g_{\uparrow\downarrow}(\mathbf{q}, \mathbf{q}', \tau)|_{\mathbf{q}'\neq-\mathbf{q}} = 0$, implying the absence of any correlation. In the case of equal but opposite momenta, we find that

$$g_{\uparrow\downarrow}(\mathbf{q}, -\mathbf{q}, \tau) = |m_{\mathbf{q}}(\tau)|^2/n_{\mathbf{q}}(\tau) = 1 - n_{\mathbf{q}}(\tau) < 1. \quad (8)$$

This corresponds to the maximum degree of correlation—as a consequence of Eq. (6), except when $n_{\mathbf{q}}(\tau) = 1$, in which case $g_{\uparrow\downarrow}(\mathbf{q}, -\mathbf{q}, \tau)$ coincides with the uncorrelated level. For bosons, the respective pair correlation is given by $g_{\uparrow\downarrow}(\mathbf{q}, -\mathbf{q}, \tau) = 1 + n_{\mathbf{q}}(\tau)$, which increases with $n_{\mathbf{q}}(\tau)$ and always stays above zero.

In order to make a better connection with the experiments at JILA [1] we note that the correlation measurements were made using absorption images after a time-of-flight expansion. This corresponds to analyzing the spatial column densities which involve integration of the 3D density along the direction of propagation of the imaging laser. Accordingly, we now analyze the momentum space analog of this procedure and calculate the correlation between momentum column density fluctuations.

The atom number operator corresponding to a z-integrated momentum column density is given by $\hat{\bar{n}}_{\mathbf{p}\sigma} = \sum_{q_z}\hat{n}_{\mathbf{q}\sigma}$, where $\mathbf{p} \equiv (q_x, q_y)$ is the reduced 2D momentum. Using Eq. (4), the average column density is found via $\bar{n}_{\mathbf{p}}(\tau) = \langle\hat{\bar{n}}_{\mathbf{p}\sigma}(\tau)\rangle = \sum_{q_z}n_{\mathbf{q}}(\tau)$ and is the same for the two spin states. Snapshots of $\bar{n}_{\mathbf{p}}(\tau)$ at different times τ are plotted in Fig. 2. The last two frames show a clear ring structure around the central background, which is consistent with the observed absorption images [1] of spatial column densities after free expansion.

The correlation function between momentum column density fluctuations in the two spin states, which we denote via $\bar{g}_{\uparrow\downarrow}(\mathbf{p}, \mathbf{p}', \tau)$, is defined as in Eq. (7) except that the operators $\hat{n}_{\mathbf{q}\sigma}$ are replaced by $\hat{\bar{n}}_{\mathbf{p}\sigma}$. In this case, the bars above the operators signify the procedure of summation
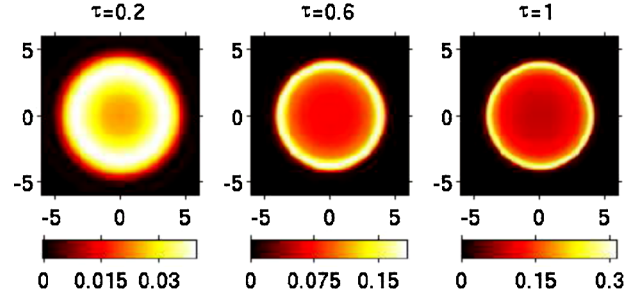

FIG. 2 (color online).   Snapshots of the average column density in momentum space $\bar{n}_{\mathbf{p}}(\tau)/l$ at different times τ, for $\delta = -16$.

over the q_z component, before taking the ensemble average. For equal but opposite momenta, $\mathbf{p}' = -\mathbf{p}$, we find (see Fig. 3) that

$$\bar{g}_{\uparrow\downarrow}(\mathbf{p}, -\mathbf{p}, \tau) = 1 - \sum_{q_z}[n_{\mathbf{q}}(\tau)]^2/\bar{n}_{\mathbf{p}}(\tau) < 1. \quad (9)$$

For any other pair of momenta ($\mathbf{p}' \neq -\mathbf{p}$), the correlation function is simply zero, implying the absence of any correlation. In the case of dissociation into bosonic atoms, the same correlation function is given by $\bar{g}_{\uparrow\downarrow}(\mathbf{p}, -\mathbf{p}, \tau) = 1 + \sum_{q_z}[n_{\mathbf{q}}(\tau)]^2/\bar{n}_{\mathbf{p}}(\tau) > 1$.

It is important to point out that the degree of correlation between atom pairs with opposite spins and momenta obtained in this model is maximal at any given density. The notion of maximal is defined here to correspond to perfect (100%) noise reduction of the number-difference fluctuations below the shot-noise level.

This can be easily understood at the level of just two modes $\hat{c}_{\mathbf{q}\uparrow}$ and $\hat{c}_{-\mathbf{q}\downarrow}$, which we define via $\hat{c}_1$ and $\hat{c}_2$. Considering the normalized variance of the particle number-difference fluctuations, $V = \langle[\Delta(\hat{n}_1 - \hat{n}_2)]^2\rangle/SN$, where SN is the shot-noise level, one can show that in the simplest case of $\langle\hat{n}_1\rangle = \langle\hat{n}_2\rangle$, the variance V and the pair correlation $g_{12} = \langle\Delta\hat{n}_1\Delta\hat{n}_2\rangle/\sqrt{\langle\hat{n}_1\rangle\langle\hat{n}_2\rangle}$ are related by $V = 1 - 2g_{12}\langle\hat{n}_1\rangle/SN$. Here the shot-noise level for fermions is given by $SN = \sum_i\langle(\Delta\hat{n}_i)^2\rangle = \sum_i\langle\hat{n}_i\rangle(1 - \langle\hat{n}_i\rangle)$, which we
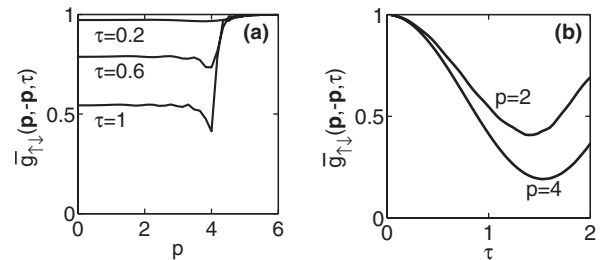

FIG. 3.   (a) Correlation between momentum column density fluctuations, $\bar{g}_{\uparrow\downarrow}(\mathbf{p}, -\mathbf{p}, \tau)$, as a function of the absolute 2D momentum $p = |\mathbf{p}|$ at different times τ, for $\delta = -16$. The correlation is larger initially, in the few-particle regime, and decreases as the number of atoms grows. Similar relationship is seen within the low-density tails and the higher-density central part of the momentum distribution. (b) Correlation signal as a function of time, for two different values of the p.

point out is always sub-Poissonian and is independent of the state of the fermion system. For a nonzero SN, the assumption of a perfect noise reduction below the shot-noise level, $V = 0$, can be used to identify the maximum degree of correlation, giving $g_{12}^{(max)} = 1 - \langle \hat{n}_1 \rangle$. This is exactly as obtained in the present model, Eq. (8). At times when the average occupancies approach one, the shot-noise itself vanishes. Therefore, the notions of sub-shot-noise fluctuations and maximal correlation become meaningless for $\langle \hat{n}_i \rangle = 1$.

For comparison, in the bosonic case the shot-noise level corresponds to that of a coherent state, $SN = \langle \hat{n}_1 \rangle + \langle \hat{n}_2 \rangle$, and never vanishes. For equal mode occupancies, it gives $V = 1 + \langle \hat{n}_1 \rangle - g_{12}$, and therefore $V = 0$ implies that $g_{12}^{(max)} = 1 + \langle \hat{n}_1 \rangle$. This is always larger than the uncorrelated level of 0, and again agrees with the actual solution to the problem of molecule dissociation.

In order to apply the results of the present uniform model to realistic trapped condensates, we remark that the quantization length L should be matched to the characteristic size of the molecular BEC. In addition, one has to ensure that the time window for dissociation is chosen such that the momentum kick $k_0 \simeq \sqrt{2m|\Delta|/\hbar}$ imparted on the atoms is not too large, so that the atoms created mostly near the trap center remain within the molecular BEC while the dissociation is on. This implies that the present results are applicable to $t \lesssim t_{max} = Lm/(2\hbar k_0)$ or $\tau \lesssim \tau_{max} = l/(4\sqrt{|\delta|})$. Considering a typical set of parameters, with 3×10^5 initial number of molecules and $l \simeq 46$ [29], the example presented here for $\delta = -16$ would produce $\sim 1.5 \times 10^4$ atoms in each spin state at $\tau = 0.6$ which compares favorably with $\tau_{max} \simeq 3$. This corresponds to 5% conversion and is consistent with the use of the undepleted molecular field approximation.

In practice, the main factors which may contribute to the reduction of the correlation signal are (i) the presence of a large thermal component in the initial molecular gas, in which case the thermal center-of-mass momenta may no longer be negligible compared to the momentum kick of the atoms k_0, and (ii) atom-atom s-wave scattering between the two spin components, which at high densities may substantially redistribute the momenta over the s-wave scattering spheres and spoil the correlations.

In summary, we have analyzed short time dynamics of dissociation of a BEC of molecular dimers into correlated fermionic atoms in two different spin states. The pair correlations between atoms with opposite spins and momenta calculated here correspond to the maximum possible degree of correlation and serve as upper bounds for more detailed calculations and comparisons with experiments. The system may find applications in precision measurements beyond the shot-noise level, as well as for fundamental tests of quantum mechanics with macroscopic number of fermions, such as demonstrations of Bohm's version of the Einstein-Podolsky-Rosen paradox [19] and tests of Bell's inequalities for spin observables.

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