Spin-exchange collisions in hot vapors creating and sustaining bipartite entanglement

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Spin-exchange collisions in alkali or alkali-noble gas vapors are at the basis of quantum sensing, nucleon structure studies, tests of fundamental symmetries, and medical imaging. We here show that spin-exchange collisions in hot alkali vapors naturally produce strong bipartite entanglement, which we explicitly quantify using the tools of quantum information science. This entanglement is shown to have a lifetime at least as long as the spin-exchange relaxation time, and to directly affect measurable spin noise observables. This is a formal theoretical demonstration that a hot and dense atomic vapor dominated by random spin-exchange collisions can support long-lived bipartite and possibly higher-order entanglement.

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Atomic spin-exchange collisions are at the basis of far-ranging explorations, from nuclear physics [1] and astrophysics [2,3] to quantum sensing [4] and medical imaging [5,6]. Spin-exchange collisions in hot alkali vapors underlie the dynamics of optical pumping and spin relaxation, both being central to producing and probing nonequilibrium magnetic substate populations by light [7–9]. More recently, the intricate properties of spin-exchange collisions [10,11] spurred the development of ultrasensitive atomic magnetometers [12–14], advancing precision tests of fundamental physics [15–17] and biomagnetic imaging [18–22]. Since relaxation is intimately connected to fluctuations, spin-exchange collisions also generate spin noise, spontaneous fluctuations of the collective spin addressed by spin noise spectroscopy [23–30].

The existing theoretical treatment [8,11,31] of alkali-metal atom spin-exchange collisions treats the two atoms emerging from a binary collision as uncorrelated, and thus accounts for single-atom observables in an atomic vapor understood as consisting of uncorrelated atoms. This approach has been quite successful, because so far experiments probed mostly single-atom observables. However, recent years witnessed the exploration of collective quantum states in such vapors. In particular, measurement-induced multiatom entanglement in hot alkali vapors was recently observed [32]. Moreover, quantum correlations in alkali or alkali-noble gas vapors were theoretically discussed in [33–35].

Here we address the quantum foundations of alkali-alkali spin-exchange collisions and show that they produce strong bipartite (atom-atom) entanglement. We formally quantify this entanglement with the tools of quantum information science, and show it persists for at least another ten spin-exchange collision times. Hence we provide a formal demonstration, using the full alkali-metal atom density matrix and the full spin-exchange interaction, that random spin-exchange collisions in a hot and dense vapor fundamentally allow for long-lived two-body entanglement. It is conceivable that these results hold true also for multibody entanglement produced by a sequence of binary spin-exchange collisions. Here we terminate this sequence at the third collision partner, limiting this discussion to bipartite entanglement. We finally show how this entanglement can be revealed through binary spin correlations affecting measurable spin noise variances.

Spin-exchange (SE) collisions between two atoms, $A$ and $B$, result from the different potential curves [36] of the singlet and triplet total spin of the colliding partners. If $s_A$ and $s_B$ are the electron spins of the colliding atoms, the singlet and triplet projectors are $P_S = \frac{1}{2} - s_A \cdot s_B$ and $P_T = \frac{1}{2} + s_A \cdot s_B$, respectively [37]. Introducing the exchange operator $P_x = P_T - P_S$, the SE interaction potential is written as [11] $V_{SE} = V_0 + V_1 P_x$.

Only the latter term drives the spin state evolution, expressed by the unitary operator $U_{SE}^{\phi} = e^{-i\int dt V_{SE}}$, where $\phi = \int dt V_1$ is the SE phase [38]. Noting that $P_x^2 = 1$, we find $U_{SE}^{\phi} = \cos \phi \mathbb{1} - i \sin \phi P_x$. Now let two uncorrelated atoms $A$ and $B$ enter a collision in the combined state $\rho_0 = \rho_A \otimes \rho_B$.

The next step in the standard derivation of SE relaxation [8,11,31] is to trace out atom $B$ ($A$) in order to find the postcollision state of atom $A$ ($B$), writing the combined postcollision state as $\rho_0' \otimes \rho_0''$, where $\rho_0' = \text{Tr}_B[\rho]$ and $\rho_0'' = \text{Tr}_A[\rho]$. With the postcollision state written as a tensor product of uncorrelated states, this approach is well suited for treating single-atom observables in a vapor described as consisting of uncorrelated atoms. Hence there is no need to keep track of the tensor-product notation and one is left with the single-atom density matrices (for single-species vapors one sets $a = b$ and omits the atom indices altogether).

We will now extend this treatment and unravel the bipartite entanglement in the postcollision two-atom state $\rho$ resulting from the action of $U_{SE}^{\phi}$ on the initial state $\rho_0$. To facilitate this...
We will first find a formal upper bound to the entanglement of $\rho$, which bound is independent of the SE phase $\phi$ and the particular colliding states $\rho_a$ and $\rho_b$. This will serve both as an indicative measure of entanglement and as a consistency check for the numerical calculations following suit and demonstrating that several colliding states of practical interest lead to significant entanglement, in cases saturating the upper bound. Incidentally, a general lower bound other than the trivial one (zero) cannot be given, since the entanglement of $\rho$ depends on $\phi$, and for $\phi = 0$ or $\phi = \pi$ it is $\rho = \rho_0$, in which case $\rho$ has zero entanglement.

We first note that the first line in Eq. (1) is a separable density matrix, i.e., it is written as $\sum_\rho \rho_\rho \otimes \rho_\rho$, with $\sum_\rho = 1$. The form $\rho^{\rho_\rho} \otimes \rho^{\rho_\rho}$ is obvious in the first term of the first line in Eq. (1). Regarding the second term, each term in the sum over $j$ multiplied by $\frac{1}{4} \sin^2 \phi$ is a physical tensor-product density matrix, because it is the result of acting on $\rho_a \otimes \rho_b$ with a completely positive map consisting of $\sigma_a$ and $\sigma_b$. The form $\sigma_a \otimes \sigma_b = \sigma_a \otimes \sigma_b$ is obvious. For example, the term $j$ results from acting on $\rho_a \otimes \rho_b$ with $M_j = \sigma_a \otimes \sigma_b$, i.e., from the operation $M_j \rho_a \otimes \rho_b M_j$. Since all $M_j$ operators are local, the resulting density matrices in each of the three such terms ($j = x, y, z$) are again of the separable form. Now, the second line in Eq. (1) is Hermitian and traceless. If this second line was absent, the density matrix $\rho$ would be separable. But as is, it generally exhibits bipartite entanglement.

Negativity is an entanglement measure [40] for bipartite systems, defined by $N(\rho) = (\|\rho^{\rho_\rho}\| - 1)/2$, where $\rho$ is the partial transpose (PT) of $\rho$. The partial transpose of a bipartite density matrix $\rho = \sum_{i,j,k,l} \rho_{ijkl} |i\rangle \otimes |j\rangle \otimes |k\rangle \otimes |l\rangle$ is $\rho^{\rho_\rho} = \sum_{i,j,k,l} \rho_{ijkl} |j\rangle \otimes |i\rangle \otimes |k\rangle \otimes |l\rangle$, i.e., the operator in the right position of the tensor product (party $B$) is transposed. The trace norm $\|\rho^{\rho_\rho}\| = \text{Tr}(\sqrt{(\rho^{\rho_\rho})^\rho})$ is a special case ($p = 1$) of the so-called Shatten-$p$ norm. Since the PT of $\rho$ in Eq. (1) is Hermitian, $\|\rho^{\rho_\rho}\|$ equals the sum of the absolute values of the eigenvalues of $\rho^{\rho_\rho}$.

To analytically calculate $\|\rho^{\rho_\rho}\|$ in all generality presents an insurmountable difficulty. However, we can calculate an upper bound to $\|\rho^{\rho_\rho}\|$, using the triangle inequality $\|A_1 + A_2 + \ldots + A_n\| \leq \|A_1\| + \|A_2\| + \ldots + \|A_n\|$, and the fact that for any constant $c$ it is $\|cA\| = |c|\|A\|$. First we note that $\sigma_a^\rho = \sigma_a$, $\sigma_b^\rho = -\sigma_b$, and $\sigma_a = \sigma_b$. It follows that the effect of the PT operation on the first line of Eq. (1) is just to change $\rho_\rho$ into $\rho_\rho^\rho$. However, $\rho_\rho^\rho$ is also a physical density matrix having opposite phases compared to $\rho_\rho$. Hence the PT, the first line is still a physical density matrix of unit trace, and hence its trace norm is 1.

For the terms in the second line of Eq. (1) we will use the identity $\|A_1 \otimes \cdots \otimes A_n\| = \|A_1\| \cdots \|A_n\|$, and the fact that the trace norm is unitarily invariant, i.e., $\|A\| = \|UAV\|$ for unitary $U$ and $V$ [41]. We appropriately choose $U$ and $V$ to be some Pauli operator $\sigma$, such that we rid all terms in the PT version of the second line of Eq. (1) from the $\sigma$ operators, also using the fact that $\sigma_\rho = \frac{1}{2} \sin 2\phi$. For example, take the term $A = \sigma_a \rho_a \otimes \sigma_b \rho_b \sigma_c$. Its partial transpose is $A^{\rho_\rho} = -\sigma_a \rho_a \otimes \sigma_b \rho_b \sigma_c \sigma_b$. The trace norm of $A^{\rho_\rho}$ is $\|A^{\rho_\rho}\| = \|\sigma_a \rho_a \otimes \sigma_b \rho_b \sigma_c \sigma_b\|$. For the first term in this product we take $U = \sigma_a$ and $V = \sigma_b$, while for the second we choose $U = \sigma_c$, and $V = \sigma_a$. Hence $\|A^{\rho_\rho}\| = \|\rho_a \otimes \rho_b \sigma_c \|= 1$. We thus reduce all terms to expressions having unit trace norm. There are 12 such terms in the expression multiplied by $(1/4) \sin^2 \phi$, and 6 such terms in the one multiplied by $-(i/4) \sin 2\phi$. Thus the negativity of $\rho$ given by Eq. (1) is bounded by

$$N(\rho) \leq \frac{3}{2} \sin^2 \phi + \frac{3}{4} |\sin 2\phi|.$$ (2)

We will next show numerically that states of experimental relevance lead to significant negativities, in cases saturating the bound of Eq. (2). We have performed an exact simulation for a $^{87}$Rb vapor (nuclear spin $I = 3/2$, 8-dimensional Hilbert space, 64-dimensional tensor product space). We use random precollision states $\rho_a \otimes \rho_b$ and a random SE phase $\phi$. Writing down the most general random, Hermitian and positive-semidefinite eight-dimensional matrix is not trivial [42]. Therefore we use random coherent superpositions of the $(FM)$ basis states to create the most general random pure states $|\psi_a\rangle$ and $|\psi_b\rangle$ [43].

The result is shown in Fig. 1(a). The largest negativity is produced for collisions of $|20\rangle$ with $|20\rangle$ and $|10\rangle$ with $|10\rangle$, both of which saturate the bound for $\phi = \pi/2$. For those cases, which are relevant to frequency standards [44], we can find the exact result $N_{\text{pt}} = \frac{3}{2} (\sin^2 \phi + |\sin \phi|\sqrt{1 + 3 \cos^2 \phi})$ [dashed upper blue line in Fig. 1(a)].

In contrast, collisions between the stretched states $|22\rangle$ with $|22\rangle$, and $|22\rangle - |22\rangle$ with $|22\rangle - |22\rangle$ produce zero negativity, $N_{\text{stretched}} = 0$. This is because stretched states are invariant under SE, so an initially uncorrelated product state of stretched states will remain invariant and uncorrelated [such zeros are not apparent in Fig. 1(a) because it is improbable, within the 5000 points, that both colliding random pure states happen to be the same stretched state].

Relevant to a highly spin-polarized vapor are mostly collisions between $|22\rangle$ and $|21\rangle$ states. In fact, such collisions produce the ubiquitous Zeeman frequency shift. Indeed, a perfectly spin-polarized vapor in the stretched state $|22\rangle$ is invariant under SE, hence there is neither any entanglement nor any Zeeman shift produced. Considering an imperfectly polarized vapor with a population of $|22\rangle$ significantly larger than the population of $|21\rangle$, it is collisions between $|22\rangle$ and $|21\rangle$ states that dominate the shift, since collisions of $|21\rangle$ with
\[ \sin^2 |\phi| = \frac{1}{2} \sin \phi |\sqrt{3} + \cos \phi |. \]

Based on Fig. 1(a) and the exact results \( N_{\text{up}} \) and \( N_{\text{st}} \), it appears that the entanglement produced by strong SE collisions, the phase of which is such [11] that the collisional average \( \sin^2 \phi \approx 1 \), is rather significant. We next turn to explicitly quantify the lifetime of this entanglement. To this end, we will bring into the picture a third atom, and consider the uncorrelated initial state \( \rho_0 \otimes \rho_0 \otimes \rho_c \). We let atoms \( A \) and \( B \) collide with phase \( \phi \) as before, and then have atom \( C \) collide with atom \( A \) with phase \( \chi \). We then trace out atom \( C \), and find the negativity of the resulting \( A-B \) state

\[
\rho' = \text{Tr}_C \{ U_C^{\dagger} U_B^{\dagger} \rho_0 \otimes \rho_0 \otimes \rho_c U_B U_C \}. \tag{3}
\]

From the resulting expression we can ignore terms proportional to either \( \sin 2\phi \) or \( \sin 2\chi \), the collisional averages of which express the collisional frequency shift and thus are very small [11], and thus we get

\[
\rho' \approx \cos^2 \chi \rho + \cos^2 \phi \sin^2 \chi \rho_0 \otimes \rho_0 \otimes \rho_c \text{Tr}_C \{ P^{\text{BC}}_e \rho_0 \otimes \rho_0 \otimes \rho_c P^{\text{BC}}_e \} + \sin^2 \phi \sin^2 \chi \text{Tr}_C \{ P^{\text{BC}}_e \rho_0 \otimes \rho_0 \otimes \rho_c P^{\text{BC}}_e \}. \tag{4}
\]

In Fig. 1(b) we plot the exact ratio \( N(\rho)/N(\rho_0) \) as a function of \( \chi \). It is seen that \( N(\rho)/N(\rho_0) \approx \cos^2 \chi N(\rho_0) \), i.e., the negativity of \( \rho' \) is approximately given by considering just the first term in Eq. (4). Since a binary SE collision happens every time interval \( T \), related to \( T_{\text{se}} \) by [8] \( \sqrt{\chi} / T = 1 / T_{\text{se}} \), and since just one \( B-C \) collision reduces the \( A-B \) negativity by \( \cos^2 \chi \), we can write

\[
\frac{dN(\rho)}{dt} \approx \frac{N(\rho') - N(\rho)}{T} \approx -\frac{N(\rho)}{T_{\text{se}}}. \tag{5}
\]

Thus the negativity \( N(\rho) \) is predicted to decay exponentially with time constant \( T_{\text{se}} \). Indeed, this is explicitly shown in the example of Fig. 1(c). To produce this plot we let both atoms \( A \) and \( B \) initially collide in the state |20⟩ with phase \( \pi / 2 \), thus producing a highly entangled state with negativity \( 3 / 2 \). We then let either \( A \) or \( B \) collide with an atom \( C \) randomly chosen among the set of states |2m⟩, and with phase sampled from a Cauchy distribution having zero mean and scale \( 10.0 \), producing an average \( \sin^2 \chi \approx 0.5 \). We consider in total 15 collisions labeled by \( n = 0, 1, \ldots, 14 \) (\( n = 0 \) is the initial \( A-B \) collision and the rest are \( A-C \) or \( B-C \) collisions). After each \( A-C \) or \( B-C \) collision atom \( C \) is traced out. We repeat this process 100 times and plot the resulting average \( \overline{N}(\rho) \), as a function of SE collision number \( n \). We find a decay “time” (in terms of the number of SE collisions) very close to \( 1 / \sin^2 \chi \) as determined from the Cauchy distribution of the \( \chi \) values. In particular, it is observed that significant negativity (\( \geq 0.1 \)) survives for about \( 11 \) times. It should be stressed that in all of the above considerations we have not specified the precise physical process realizing the tracing out of atom \( C \). But our starting point was the existing single-atom derivation tracing out atom \( B \) (and \( A \)) when obtaining the single-atom density matrix under the assumption of instant \( A-B \) decorrelation. We found the natural

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**FIG. 1.** (a) Negativity of \( \rho \) in Eq. (1). Red solid line is the upper bound of Eq. (2). The 5000 orange dots correspond to random pure initial states \(|\phi_a⟩\) and \(|\phi_b⟩\) for \(^{87}\text{Rb}\) and random SE phase \( \phi \). Upper (lower) dashed blue lines are the analytic negativities \( N_{\text{up}} \) (\( N_{\text{st}} \)) for collisions of |20⟩ with |20⟩ (|22⟩ with |21⟩). Dotted curve at zero corresponds to collisions between stretched states, |22⟩ with |22⟩ and |2 − 2⟩ with |2 − 2⟩. (b) Ratio of \( N(\rho') / N(\rho_0) \) for the same states corresponding to the orange dots in (a), where \( \rho' \) results from a \( B-C \) collision with random phase \( \chi \) and random \( C \)-atom state \(|\phi_c⟩\), after tracing out atom \( C \). Solid line is \( \cos^2 \chi \). (c) Evolution of \( A-B \) negativity (blue dots), starting out from a highly entangled \( A-B \) state with negativity 1.5, then \( A \) or \( B \) colliding with \( C \) atoms randomly chosen among |2m⟩ with random phase \( \chi \). Red solid line is an exponential decay with constant \( n_0 = 2.3 \), close to \( 1 / \sin^2 \phi \approx 2.0 \) resulting from the Cauchy distribution of \( \chi \) having center 0.0 and scale 10.0.
timescale of this decorrelation by going into a deeper layer
of the many-body spin dynamics and invoking the interac-
tion with a third atom \( C \). In fact, we forcefully and instantly decor-
correlations with the third collision partner (atom \( C \)).
However, it is expected that atom \( C \) will gradually (through
further collisions) extract information from the \( A-B \) state
[45–47], rendering \( T_{\text{end}} \) a lower bound for the \( \tau \) entanglement
time.

Concluding, we outline how the bipartite entanglement con-
sidered herein can manifest itself experimentally. Consider a
spectroscopic measurement of the collective spin of \( N \) atoms,
\( \mathcal{F}_q = \sum_{j=1}^{N} f_{j}^q \), where \( f_{j}^q \) is the \( q \) component of the \( j \)-th atom
total spin, with \( q = x, y, z \). The variance of \( \mathcal{F}_q \) is in principle
readily measurable, and is given by \((\Delta \mathcal{F}_q)^2 = (\mathcal{F}_q^2) -
(\mathcal{F}_q)^2 = \sum_{j=1}^{N}(\Delta f_{j}^q)^2 + \sum_{\{q\}} C_{ij}^{\text{ij}}\), where \( C_{ij}^{\text{ij}} = (\langle f_{i}^q f_{j}^q \rangle -
\langle f_{i}^q \rangle \langle f_{j}^q \rangle) \). Clearly, for uncorrelated atoms it is \( C_{ij}^{\text{ij}} = 0 \), and
the total variance equals the sum of the individual atom vari-
ances. Now, it is seen that a nonzero \( C_{ij}^{\text{ij}} \) is connected with
the entanglement produced by an SE collision between atoms
\( A \) and \( B \). Indeed, using the postcollision state \( \rho \) of Eq. (1) we
can find both terms entering \( C_{ij}^{\text{ij}} \). It is \( \langle f_{i}^q f_{j}^q \rangle = \text{Tr}(\rho f_{i}^q \otimes
f_{j}^q) \), \( \langle f_{i}^q \rangle = \text{Tr}(\rho f_{i}^q) \), with \( \rho_{a} = \text{Tr}_{A}[\rho] \) and
\( \rho_{b} = \text{Tr}_{B}[\rho] \). In Fig. 2 we plot examples of \( C_{ij}^{\text{ij}} \) for
collisions within the \( F = 2 \) manifold. It is evident that a large
negativity \( N(\rho) \) is connected with a large \( |C_{ij}^{\text{ij}}| \).

To show the effect of \( C_{ij}^{\text{ij}} \) on spin variances we consider
the following three cases of practical interest (in all examples
we take \( \phi = \pi/2 \)): (i) A state having Zeeman coherence, e.g.,
\( |\psi\rangle = (|22\rangle + |21\rangle)/\sqrt{2} \). After SE, the resulting variance of
\( \mathcal{F}_z \) for this state is 3.5, of which 21\% comes from positive
binary correlations. (ii) A state exhibiting alignment, e.g.,
\( |\psi\rangle = (|22\rangle + |2-2\rangle)/\sqrt{2} \). Here the resulting variance of
\( \mathcal{F}_z \) is 8, of which 62.5\% is the contribution of positive correla-
tions. (iii) The state \( |20\rangle \) of interest to clock transitions. Here
the resulting variance of \( \mathcal{F}_z \) is zero. This is due to negative cor-
relations [see Fig. 2(b)] completely canceling the variance’s
correlated contribution. For yet another example, a mea-
surement of the projector to the lower \( F = 1 \) manifold results
in a variance of 0.75, of which 37.6\% comes from correla-
tions. Many other scenarios can similarly lead to significant
effects on spin variances due to correlations established by
the entanglement spontaneously produced by SE collisions.
Such correlations will have to be fully understood in order
to benchmark any metrological improvement of entangled
states produced by external means (e.g., interaction with light,
as in [32]) against the baseline variances determined by the
underlying collisional physics.

Concluding, we have explored the atom-atom entangle-
generated by spin-exchange collisions in hot alkali
vapors. Our results should be equally applicable to alka-
noble gas collisions, and have the potential to further advance
the understanding and design of nontrivial collective quantum
states for use in quantum technology.

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\textit{Correction:} The curve on the left-hand side in the previously published Figure 1(b) was processed improperly during the production stage and its rendition has been fixed.