Entanglement and spin squeezing of Bose condensed atoms

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(Dated: November 19, 2018)

We propose a direct, coherent coupling scheme that can create massively entangled states of Bose-Einstein condensed atoms. Our idea is based on an effective interaction between two atoms from coherent Raman processes through a (two atom) molecular intermediate state. We compare our scheme with other recent proposals for generation of massive entanglement of Bose condensed atoms. We also present explicit calculations that support detecting maximally entangled states without requiring single atom resolved measurements.

I. INTRODUCTION

Entanglement lies at the heart of the difference between the quantum and classical multi-particle world. It is the phenomenon that enables quantum information and quantum computing with many qubits. Recently, several interesting developments have occurred in studies of massively entangled atomic states. Based on the proposals of Sorensen and Molmer \cite{1}, a controlled, entangled state of 4−ions was successively created by the NIST ion trap group \cite{3}. Zeilinger and coworkers prepared three entangled photon or GHZ states by selecting from two beams of entangled photon pairs \cite{4}. Entanglement between two atoms and a microwave photon were also detected in a “step-by-step” process \cite{4}.

Of these and other related developments, the idea of Molner and Sorensen \cite{1} is especially interesting. In their scheme a direct coupling to the multiparticle, entangled final state was made possible through a virtual, intermediate state which was a common (quantum) mode of the motion of all the ions. Similar type interactions were also obtained by Milburn \cite{5}. Both proposals allow for creation of massive entangled states by unitary evolution, starting from certain pure initial states.

A Bose-Einstein condensate is a convenient source of atoms in initially pure and separable states. Zoller and coworkers suggested creating massive entangled spin squeezed states from a two component condensate using the inherent atom-atom interactions \cite{6} and further investigated schemes to dynamically create many particle entangled states of a two component BEC \cite{7}. Spin exchange interaction of a spinor condensate was also proposed as a candidate for creating entangled pairs of atoms \cite{8, 9}.

Most of these proposals work in the two mode approximation where one motional state is assumed for each spinor component of condensed states. Recently, Sorensen demonstrated the validity of this method within Bogoliubov theory \cite{10}. Similar studies were previously performed, extensively, for condensate atoms in a double well potential \cite{11, 12, 13, 14, 15, 16}.

Recently, two of us proposed a new type of atom-atom coupling that achieves optimal spin squeezing in two mode Bose condensed atoms \cite{17}. In this study we provide further details of our proposal and compare its potential for both the creation of massive entangled states \cite{18, 19} and improved spin squeezing \cite{8, 9, 18, 19}.

This paper is organized as follows. First, we briefly review the Raman process with photon momentum transfer for a single three-level Λ-type atom. We then extend the analysis to the case of two atoms, using intermediate molecular states. The resulting effective interaction is then compared with models in several other recent studies of creating entanglement with Bose condensed atoms. We then provide an explicit calculation of the time reversed dynamics to confirm the generation of maximally entangled states without requiring single atom resolved measurements \cite{20, 21}. We conclude with a careful analysis of the limitations and advantages of our model.

II. SINGLE ATOM, TWO-PHOTON PROCESS THROUGH AN ATOMIC INTERMEDIATE STATE

We consider a three level Λ-type atom described by the Hamiltonian

\begin{equation}
H = \sum_{\mu=g,e,g'} \left[ \frac{\vec{p}^2}{2M} + V_\mu(\vec{r}) + \hbar \omega_{\mu g} \right] |\mu\rangle \langle \mu| + H_{\text{int}}, \tag{1}
\end{equation}

where the dipole interaction term is

\begin{equation}
H_{\text{int}} = -d_{eg} \cdot \vec{E}_1(\vec{r}) e^{i\vec{k}_1 \cdot \vec{r} - i\omega_{1}t} |g\rangle \langle e| + \text{h.c.} - d_{g'g} \cdot \vec{E}_2(\vec{r}) e^{i\vec{k}_2 \cdot \vec{r} - i\omega_{2}t} |g'\rangle \langle e| + \text{h.c.} \tag{2}
\end{equation}

for the general Raman process from initial state $|g\rangle$ to final state $|g'\rangle$ through an intermediate state $|e\rangle$. $V_\mu(\vec{r})$ denotes the, possibly different, trapping potentials for the different electronic states $|\mu\rangle$ with internal energy $\omega_{\mu g} = \omega_{\mu e} - \omega_{gg}$. $\vec{E}_1(\vec{r}) e^{i\vec{k}_1 \cdot \vec{r} - i\omega_{1}t}$ denotes the running wave amplitude of the laser field.

We define $\Delta = \omega_1 - \omega_{eg}$ and $\delta = \omega_2 - \omega_1$. Without loss of generality, assuming $d_{eg} = d_{g'g} = d$, $\Omega_z = 2d \cdot \vec{E}_j/\hbar$ is the Rabi frequency for the corresponding dipole connected transition. In the limit of a resonant two photon process with large detuning from the intermediate atomic excited state $|e\rangle$, we adiabatically eliminate $|e\rangle$ to obtain...
an effective Hamiltonian

\[ \mathcal{H}_{\text{eff}} = \sum_{\mu=g,g'} \left[ \frac{P_\mu^2}{2M} + V_\mu(\vec{r}) \right] |\mu\rangle\langle\mu| + V_{\text{B/R}}, \]

with,

\[ V_B = \frac{\hbar}{4\Delta} \sum_j \Omega_j e^{i\vec{k}_j \cdot \vec{r} - i\delta_j t} \sum_j \Omega_j^* e^{-i\vec{k}_j \cdot \vec{r} + i\delta_j t} |g\rangle \langle g|, \]

\[ V_R = \frac{\hbar}{4\Delta} \Omega_1 e^{i\vec{k}_1 \cdot \vec{r} - i\omega_1 t} \Omega_* e^{-i\vec{k}_2 \cdot \vec{r} + i\omega_2 t} |g\rangle \langle g'| + \text{h.c.}, \]

where distinguish the two types Raman process according to whether the final internal state \(|g'\rangle\) is the same as or different from the initial state \(|g\rangle\), respectively. When \(|g'\rangle = |g\rangle\), we identify the general Raman process as Bragg diffraction \([21]\).

In momentum space, the above two photon process involves the simultaneous absorption of a photon \((\vec{k}_1, \omega_1)\) by an atom in state \(|g; \vec{p}\rangle\) and stimulated transition to state \(|g'; \vec{p} - \hbar\vec{k}_2 + \hbar\vec{k}_1\rangle\) with emission of a photon \((\vec{k}_2, \omega_2)\). For the case of Bragg diffraction \((g' = g)\) with two counter-propagating waves

\[ \vec{k}_1 = \vec{k}, \quad \vec{k}_2 \approx -\vec{k}, \quad \vec{K} = \vec{k}_1 - \vec{k}_2 \approx 2\vec{k}, \]

we obtain

\[ V_B(\vec{r}, t) = \frac{\hbar}{4\Delta} \left[ |\Omega_1|^2 + |\Omega_2|^2 + \Omega_1^* \Omega_* e^{i\delta_2 t} e^{i\vec{K} \cdot \vec{r}} + \text{h.c.} \right] |g\rangle \langle g|, \]

The first two terms are AC Stark shifts that can be neglected. For an atom initially in state \(|g, \vec{p}_g\rangle\), the last two terms of Eq. (1) couples to momentum states \(\vec{p}_g \pm 2\hbar\vec{k}\), respectively. By appropriately choosing \(\delta_2\) one can selectively enhance coupling to only one momentum state or side mode. The resonance condition is defined by energy conservation in the two photon process

\[ \hbar \omega_1 + \frac{\vec{p}_g^2}{2M} = \hbar \omega_2 + \frac{(\vec{p}_g + \hbar \vec{K})^2}{2M}, \]

which gives

\[ \hbar \delta_2 = \frac{\vec{p}_g^2}{2M} - \frac{(\vec{p}_g + \hbar \vec{K})^2}{2M} \approx \frac{\hbar^2 \vec{K}^2}{2M}. \]

for an atom with \(\vec{p}_g \sim 0\). Hence under conditions resonant for Bragg diffraction, the effective coupling \((\vec{p})\) creates superposition states like,

\[ \langle \psi \rangle = \alpha |g\rangle |\vec{p}_g\rangle + \beta |g\rangle |\vec{p}_g \pm 2\hbar\vec{k}\rangle. \]

For Bose-condensed atoms, the above discussion still applies to any of the identical single atoms. When a condensate of \(N\) atoms is involved (initial motional state \(|p_g \sim 0\rangle\)), one simply creates a condensate in state \(|10\rangle\), which is not an entangled state because it is simply putting \(N\) atoms into the same state \(|10\rangle\), i.e.

\[ \left( a_\psi^\dagger \right)^N \langle \text{vac} \rangle \sim \psi(\vec{r}_1)\psi(\vec{r}_2)\cdots\psi(\vec{r}_N) = |\psi\rangle^{\otimes N}. \]

In practice this can only be done if atoms are noninteracting, or that the effect of interactions is small during the time of atomic Bragg diffraction. Bragg diffraction of condensates atoms was demonstrated first by the NIST-Gaithersburg group \([20]\). Higher motional states \(\vec{K} = \pm 2m\vec{k} (m \text{ integer})\) was also obtained through, higher order, \(2m\)-photon couplings.

For the Raman process with two co-propagating waves \(\vec{k}_1 = \vec{k}_2\) involving nearly degenerate ground states \(|g\rangle\) and \(|g'\rangle\), we have

\[ V_R = \frac{\hbar}{4\Delta} \Omega_1 \Omega_*^* e^{i\delta_2 t} |g\rangle \langle g'| + \text{h.c.}, \]

where AC Stark shifts from second order processes involving the same laser fields have been neglected. The Raman resonance condition is then simply \(\omega_1 - \omega_2 = \omega_1 - \omega_2\). Note that co-propagating Raman results in an effective coupling \((\vec{p})\) which is constant, independent of the position \(r\). The two photon process then is essentially insensitive to motional effects \((\vec{k}_2 \sim \vec{k}_1)\).

III. TWO-ATOM, TWO-PHOTON PROCESS THROUGH AN INTERMEDIATE MOLECULAR STATE

We now consider a model involving two \(\Lambda\)-type atoms whose initial and final states are described by the same non-interacting atomic states. The intermediate excited state, however, is now chosen to be a bound, molecular, excited state, similar to those utilized in recent photoassociation experiments \([21]\). Neglecting configurations not directly involved in the two photon process, the two atom Hamiltonian can be written as

\[ \hbar \omega_1 + \frac{\vec{p}_g^2}{2M} = \hbar \omega_2 + \frac{\left(\vec{p}_g + \hbar \vec{K}\right)^2}{2M}, \]

with

\[ V_B = \frac{\hbar}{4\Delta} \sum_j \Omega_j e^{i\vec{k}_j \cdot \vec{r} - i\delta_j t} \sum_j \Omega_j^* e^{-i\vec{k}_j \cdot \vec{r} + i\delta_j t} |g\rangle \langle g|, \]

\[ V_R = \frac{\hbar}{4\Delta} \Omega_1 e^{i\vec{k}_1 \cdot \vec{r} - i\omega_1 t} \Omega_* e^{-i\vec{k}_2 \cdot \vec{r} + i\omega_2 t} |g\rangle \langle g'| + \text{h.c.}, \]

where distinguish the two types Raman process according to whether the final internal state \(|g'\rangle\) is the same as or different from the initial state \(|g\rangle\), respectively. When \(|g'\rangle = |g\rangle\), we identify the general Raman process as Bragg diffraction \([21]\).

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we obtain

\[ V_B(\vec{r}, t) = \frac{\hbar}{4\Delta} \left[ |\Omega_1|^2 + |\Omega_2|^2 + \Omega_1^* \Omega_* e^{i\delta_2 t} e^{i\vec{K} \cdot \vec{r}} + \text{h.c.} \right] |g\rangle \langle g|, \]

The first two terms are AC Stark shifts that can be neglected. For an atom initially in state \(|g, \vec{p}_g\rangle\), the last two terms of Eq. (1) couples to momentum states \(\vec{p}_g \pm 2\hbar\vec{k}\), respectively. By appropriately choosing \(\delta_2\) one can selectively enhance coupling to only one momentum state or side mode. The resonance condition is defined by energy conservation in the two photon process

\[ \hbar \omega_1 + \frac{\vec{p}_g^2}{2M} = \hbar \omega_2 + \frac{\left(\vec{p}_g + \hbar \vec{K}\right)^2}{2M}, \]

which gives

\[ \hbar \delta_2 = \frac{\vec{p}_g^2}{2M} - \frac{\left(\vec{p}_g + \hbar \vec{K}\right)^2}{2M} \approx \frac{\hbar^2 \vec{K}^2}{2M}. \]
\[ \mathcal{H} = \sum_{\mu = g, g'} \left[ \sum_{i=1,2} \left( \frac{p_i^2}{2M} + V_\mu(r_i) \right) + 2\hbar \omega_{\mu g} \right] |\mu, \mu \rangle \langle \mu, \mu | + \sum_{\mu = g, g'} U_{\mu\mu}(|\vec{r}_1 - \vec{r}_2|) |\mu, \mu \rangle \langle \mu, \mu | + \sum_{i=1,2} \left( \frac{p_i^2}{2M} + V_{g', g}(r_i) \right) + U_{g'g}(|\vec{r}_1 - \vec{r}_2|) + \hbar \omega_{g'g} |g', g \rangle \langle g', g | + \sum_{i=1,2} \left( \frac{p_i^2}{2M} + V_{e_i}(r_i) \right) + U_{e_i}(|\vec{r}_1 - \vec{r}_2|) + \hbar \omega_{eg} \right] |e_b \rangle \langle e_b | + H_{\text{int}}, \tag{13} \]

where states $|\mu, \mu \rangle$ for $\mu = g, g'$ denote symmetrized electronic states of two atoms, each in $|\mu \rangle$. $|g', g \rangle$ denote the symmetrized state with one in $|g \rangle$ and the other in $|g' \rangle$. $V_{\mu g, g', g, e_b}$ denote external trapping potentials in state $|\mu \rangle$, $U_{\mu\mu}$ and $U_{g'g}$ are atom-atom interactions which at large inter-atomic distances are described by the usual van der Waals type terms, and at short range modified by coulomb effects. The intermediate molecular state family, denoted by $|e_b \rangle$, can be accessed through the direct dipole coupling term $H_{\text{int}}$. $U_{e_b}$ is the Born-Oppenheimer molecular (of atom 1 and 2) binding potential for internal state manifold $|e_b \rangle$, which contain bound levels to be used as intermediate states.

Introducing center of mass $\vec{R} = (r_1 + r_2)/2$ and relative coordinate $\vec{r} = r_2 - r_1$ for the two atoms, we can express

\[ \vec{r}_1 = \vec{R} - \frac{\vec{r}}{2}, \quad \vec{r}_2 = \vec{R} + \frac{\vec{r}}{2}, \tag{14} \]

and

\[ \frac{\vec{p}_1^2}{2M} + \frac{\vec{p}_2^2}{2M} = -\frac{\vec{p}^2}{2(2M)} + \frac{\vec{p}^2}{2(M/2)}. \tag{15} \]

When $|e_b \rangle$ is asymptotically connected with $|e, \mu \rangle = (|e \rangle |\mu \rangle + |\mu \rangle |e \rangle) / \sqrt{2}$ (for $\mu = g, g'$), we can express the dipole coupling as

\[ H_{\text{int}} = -\sum_{i=1,2} \vec{a}_i \cdot \vec{E}_1 e^{i \vec{k}_1 \cdot \vec{r}_1 - i \omega_1 t} \langle g |e \rangle |e \rangle + \text{h.c.} \]

\[ -\sum_{i=1,2} \vec{a}_i \cdot \vec{E}_2 e^{i \vec{k}_2 \cdot \vec{r}_2 - i \omega_2 t} \langle g' |e \rangle |e \rangle + \text{h.c.} \]

\[ = -\frac{\hbar \Omega_1}{\sqrt{2}} \cos(\frac{\vec{k}_1 \cdot \vec{r}}{2}) e^{i \vec{k}_1 \cdot \vec{R} - i \omega_1 t} |g, g \rangle |e_b \rangle + \text{h.c.} \]

\[ -\frac{\hbar \Omega_2}{\sqrt{2}} \cos(\frac{\vec{k}_2 \cdot \vec{r}}{2}) e^{i \vec{k}_2 \cdot \vec{R} - i \omega_2 t} |g', g' \rangle |e_b \rangle + \text{h.c.}, \tag{16} \]

where we have assumed $\vec{a}_i = \vec{a}_f = \vec{a}$ for the $|\mu \rangle \rightarrow |e \rangle$ transition of atom $i$. We note that the electronic dipole coupling $\Omega_i$ will vary considerably with $\vec{r}$ for small values of $r$, due to the relatively short range of the molecular interactions.

To express the intermediate state in terms of the particular molecular resonance or bound state $|e_b \rangle$, we now take a look at the eigen-structure within the $|e_b \rangle$ family. We will essentially base our discussion on some kind of molecular state asymptotically connected to the limit of one ground $|g \rangle$ and one excited atom $|e \rangle$, just as the $\Omega^c_0$ state, extensively discussed by D. Heinz and others in their photo association work [22].

The recent experimental work on the production of ground state molecules from an atomic condensate by a two-photon Raman process [21] provides additional motivation to explore our ideas experimentally. The photoassociation process [22, 23, 24] used by Heinz et al. relies on the transition strength of going from a (two atom) ‘free’ to a (molecular) ‘bound’ state via an intermediate, excited (molecular) ‘bound’ state. What we desire, on the other hand, is a transition from a (two atom) ‘free’ to ‘free’ (relative motion) state via an intermediate, excited (molecular) ‘bound’ state. More detailed discussions about feasibility of such transitions can be given provided all molecular potential curves are available.

The question of whether this is possible or not does not seem to depend on the sample density (in the weakly interacting limit), but will depend, as we shall show below, to a large degree on the trap strength and the excited bound state structure. Assuming all trapping potentials $V_g$, $V_{g'}$, and $V_{e_b}$ to be harmonic, the separation of center-of-mass and relative coordinates for the trapping potentials is

\[ \frac{1}{2} M \sum_{j=1,2,3} \nu_j^2 x_{1j}^2 + \frac{1}{2} M \sum_{j=1,2,3} \nu_j^2 x_{2j}^2 \]

\[ = \frac{1}{2} (2M) \sum_{j=1,2,3} \nu_j^2 X_j^2 + \frac{1}{2} (M/2) \sum_{j=1,2,3} \nu_j^2 x_{1j}^2 \]

\[ = V_{tr} + V_{\text{R}}, \tag{17} \]

where $x_{ij} = \hat{e}_j \cdot \vec{r}_i$, $X_{ij} = \hat{e}_j \cdot \vec{R}$, and $x_j = \hat{e}_j \cdot \vec{r}$ with $\hat{e}_j$ the unit vector in the $j$ direction.

When short range interactions $U_{\mu\mu}(\vec{r})$ are approximated by their optical contact forms, Wilkens and coworkers provide analytic solutions for two interacting atoms inside a harmonic trap [23]. In principle, from

\[ \left[ \frac{p^2}{2(M/2)} + V_{\mu\mu}(\vec{r}) + U_{\mu\mu}(\vec{r}) \right] \phi_n^\mu(\vec{r}) = E_n^\mu \phi_n^\mu(\vec{r}), \tag{18} \]
we can find all bound states $\phi_m^b(\vec{r})$ due to the external trapping potential $U_{\mu\mu}(\vec{r})$. We note that if the range of the inter-atomic interaction $\ll r \ll$ size of the ground state of the harmonic trap, then these bound states should resemble the standard low energy scattering solutions.

Similarly, the bound state of the intermediate, excited molecular level satisfies

$$\left[ \frac{\vec{p}^2}{2(M/2)} + V_{tr}^e(\vec{r}) + U_{e_b}(\vec{r}) \right] \phi_m^e(\vec{r}) = E_m^e \phi_m^e(\vec{r}), \hspace{1cm} (19)$$

where the energy $E_m^e$ is measured from the asymptotic energy $\hbar \omega_{eg}$. The index $m$ represents the rotational and vibrational quantum numbers. For all bound states with $E_m^e < 0$, the effect of external trapping potential $V_{tr}^e$ is much less than for the free atoms, since the corresponding bound state wave function is localized inside the molecular potential well $U_{e_b}$.

Similarly, complete bound states $|l\rangle$ can also be introduced for $|g', g\rangle$ if needed. They satisfy

$$\left[ \frac{\vec{p}^2}{2(M/2)} + V_{tr}^{g', g}(\vec{r}) + U_{g', g}(\vec{r}) \right] \psi_l(\vec{r}) = E_l^{g', g} \psi_l(\vec{r}). \hspace{1cm} (20)$$

In the $(\vec{R}, \vec{r})$ basis, keeping only one intermediate bound state $m_b$, we can then write our previous Hamiltonian, Eq. (13), as

$$\mathcal{H} = \sum_{n_{\mu}} \sum_{\mu=g, g'} \left[ \frac{\vec{p}^2}{2(M/2)} + V_{tR}^\mu(\vec{R}) + 2\hbar \omega_{eg} + E_n^\mu \right] |\mu, \mu; n_{\mu}\rangle \langle \mu, \mu; n_{\mu}|$$

$$+ \sum_l \left[ \frac{\vec{p}^2}{2(M/2)} + V_{tR}^{g', g}(\vec{R}) + \hbar \omega_{e_b} + E_l^{g', g} \right] |g', g; l\rangle \langle g', g; l|$$

$$+ \left[ \frac{\vec{p}^2}{2(M/2)} + V_{tR}(\vec{R}) + \hbar \omega_{eg} + E_{m_b}^e \right] |e_b; m_b\rangle \langle e_b; m_b| + H_{int}, \hspace{1cm} (21)$$

and the interaction term Eq. (10) now becomes

$$H_{int} = -\frac{\hbar \Omega_1}{\sqrt{2}} \sum_{n_{\mu}} \eta_{n_{\mu} m_b} e^{i\vec{k}_1 \cdot \vec{R} - i\omega_1 t} |g, g; n_{\mu}\rangle \langle e_b; m_b| + h.c.$$

$$-\frac{\hbar \Omega_2}{\sqrt{2}} \sum_{n_{\mu}} \eta_{n_{\mu} m_b} e^{i\vec{k}_2 \cdot \vec{R}/2 - i\omega_2 t} |g, g'; n_{\mu}\rangle \langle e_b; m_b| + h.c., \hspace{1cm} (22)$$

where

$$\eta_{n_{\mu} m_b} = \frac{1}{|d|} \langle n_{\mu}| d(\vec{r}) \cos(\vec{k} \cdot \vec{r}/2) |m_b\rangle$$

$$\approx \frac{1}{|d|} \int d\vec{r} \phi_n^b(\vec{r}) d(\vec{r}) \cos(\vec{k} \cdot \vec{r}/2) \phi_{m_b}^e(\vec{r}), \hspace{1cm} (23)$$

which is a measure of the free-bound transition amplitude. We note that in contrast to other expressions for the free-bound transition amplitude [22, 23], the motional wave functions $|n_{\mu}\rangle$ and $|m_b\rangle$ are now normalized in the trap rather than energy or momentum normalized. Although the modification of the inner part of the trap potential for $r \leq 100 (a_0)$ due to the inter-atomic potentials is to be included, these values can be determined for selected molecular states from the results of photo-association experiments. They can also be computed directly if accurate potentials are available. Detailed discussions are given by Verhaar [22], Julienne [23] and Javaninen [24].

In general, the coupling coefficient will vary depending on intermediate state. It is therefore important to pick the largest one of them. We then perform an adiabatic elimination of the excited state by appropriate choice of laser detunings, and obtain the effective Hamiltonian,
with

\[
V_B = \frac{\hbar}{4\Delta} \sum_{n,l} \left[ \sum_j \Omega_j \eta_{lmn} e^{\imath \mathbf{k}_j \cdot \mathbf{R} - \imath \omega_j t} \right] \left[ \sum_j \Omega_j \eta_{lmn}^* e^{\imath \mathbf{k}_j \cdot \mathbf{R} - \imath \omega_j t} \right]^\dagger |g, g; n\rangle \langle g, g; l| ,
\]

\[
V_R = \frac{\hbar}{4\Delta} \sum_{n,n'} \left[ \Omega_1 \eta_{nmn} e^{\imath \mathbf{k}_1 \cdot \mathbf{R} - \imath \omega_1 t} \right] \left[ \Omega_2 \eta_{nmn}^* e^{\imath \mathbf{k}_2 \cdot \mathbf{R} - \imath \omega_2 t} \right]^\dagger |g', g'; n\rangle \langle g, g; n| + h.c. ,
\]

where we have defined \( \Delta = \omega_1 - \omega_2 - E_{n_k}^2 / \hbar \). Note the \((\sqrt{2})^2\) enhancement due to excitation to an asymptotic, symmetric state.

More generally, one can include all motional states and still be able to find the effective coupling between the selected electronic states, to obtain similar results [26].

Our analysis now follows along the same lines as in the previous case of a single atom. For example, if we consider only the lowest motional state \( n_\mu = 0 \) (a situation well approximated by Bose condensed atoms), then our Hamiltonian becomes

\[
\hat{\mathcal{H}} = \sum_{\mu = g, g'} \left[ \frac{\bar{\mathcal{P}}_\mu}{2(2M)} + V_\mu^\mu (\bar{R}) + 2\hbar \omega_\mu + E_\mu^\mu \right] |\mu, \mu; 0\rangle \langle \mu, \mu; 0| + V_{B/R},
\]

with

\[
V_B = \frac{\hbar}{2\Delta} \eta_{0m0} \eta_{0m0}^* \left[ \sum_j \Omega_j e^{\imath \mathbf{k}_j \cdot \mathbf{R} - \imath \omega_j t} \right] \left[ \sum_j \Omega_j e^{\imath \mathbf{k}_j \cdot \mathbf{R} - \imath \omega_j t} \right]^\dagger |g, g; 0\rangle \langle g, g; 0'| ,
\]

\[
V_R = \frac{\hbar}{2\Delta} \eta_{0m0} \eta_{0m0}^* \Omega_1 \Omega_2 \left[ e^{\imath \mathbf{k}_1 \cdot \mathbf{R} - \imath \omega_1 t} \right] \left[ e^{\imath \mathbf{k}_2 \cdot \mathbf{R} - \imath \omega_2 t} \right]^\dagger |g', g'; 0\rangle \langle g, g; 0'| + h.c. .
\]

For Bragg diffraction by two counter-propagating waves, the above result reduces to

\[
V_B(\bar{R}, t) = \frac{\hbar}{2\Delta} \eta_{0m0} \left[ \left| \Omega_1 \right|^2 + \left| \Omega_2 \right|^2 + \Omega_1 \Omega_2^* e^{\imath \mathbf{k}_1 \cdot \mathbf{R}} + \Omega_1^* \Omega_2 e^{-\imath \mathbf{k}_2 \cdot \mathbf{R}} \right] |g, g; 0\rangle \langle g, g; 0| .
\]

Similar to the single atom case, Bragg diffraction involves the simultaneous absorption and stimulated emission of photons, i.e. the elementary Bragg process involves a total of two photons. In contrast, however, to the single atom case, a pair of atoms are now diffracted as illustrated in Fig. 1. Hence for Bose condensed atoms with \( p_i \sim 0 \), Bragg diffraction produces atoms in the state

\[
|\psi\rangle_M = \alpha |g, g; 0\rangle |p_1 \approx 0, p_2 \approx 0 \rangle + \beta |g, g; 0\rangle e^{\pm \imath \mathbf{K} \cdot \mathbf{R}} + \alpha^* |g, g; 0\rangle |p_1 \approx 0, p_2 \approx 0 \rangle + \beta^* |g, g; 0\rangle |p_1 = h\mathbf{K}/2, p_2 = h\mathbf{K}/2 \rangle .
\]

Not surprisingly, the momentum shift is only half the value of atomic Bragg diffraction, due to the fact that only two photons are involved for the pair of atoms. The resonance condition occurs at

\[
\hbar \omega_1 + \frac{p_i^2}{2(2M)} + \frac{p_j^2}{2(M/2)} = \hbar \omega_2 + \frac{p_i^2}{2(2M)} + \frac{p_j^2}{2(M/2)} ,
\]

\[
\hbar \delta_2 = \frac{P_i^2}{2(2M)} - \frac{(\mathcal{P}_i + h\mathbf{K})^2}{2(2M)} \approx - \frac{h^2 \mathbf{K}^2}{2(2M)} ,
\]

i.e. half the atomic Bragg resonance (3). It is worth mentioning that for a condensate with many atoms, the state obtained from diffraction via a two atom intermediate state will be complicated. It is generally not of the form \( |a_{(0,0)}^1\rangle |\text{vac}\rangle \).

For a nearly degenerate Raman process with two co-propagating waves, we obtain for atoms initially at rest,

\[
V_R(\bar{R}, t) = \frac{\hbar}{2\Delta} \eta_{0m0} \eta_{0m0}^* \Omega_1 \left[ e^{\imath \mathbf{k}_1 \cdot \mathbf{R}} |g, g; 0\rangle \langle g, g; 0'| + h.c. \right.
\]

\[
+ \left. \Omega_1^* e^{-\imath \mathbf{k}_2 \cdot \mathbf{R}} |g, g; 0\rangle \langle g, g; 0'| + h.c. \right] .
\]
The resonance condition now becomes $\omega_1 - \omega_2 = 2\omega_{g'g}$, i.e. the Raman process changes each atom’s state ($g \rightarrow g'$) and so each atom acquires the energy deficit $\hbar \omega_{g'g}$. Note that there is no $\vec{R}$ dependence of the coupling coefficient in this case.

If the Raman process is arranged to allow for simultaneous scattering to two different final states $|g'\rangle$ and $|g''\rangle$, the coherent coupling becomes,

$$ V_{R}^{[2]}(\vec{R},t) = \frac{\hbar \Omega_{R}}{2} e^{i\delta t} |g, g; 0\rangle \langle g', g'; 0'\rangle + h.c. $$

$$ + \frac{\hbar \Omega_{R}'}{2} e^{i\delta' t} |g, g; 0\rangle \langle g'', g'''; 0''\rangle + h.c. $$

Such a coupling can be obtained among different Zeeman levels in the atomic ground state.

Finally, we note that our discussions above can also be applied to a pair of different species of atoms, e.g. using molecular intermediate state from Li-Cs dimer could create entangled pairs of Li and Cs. We also note that to be formally correct, the second order perturbation process also needs to be considered for the state $|g', g\rangle$, which should also have some type of effective coupleings with itself and as well as with the states $|g, g\rangle$ and $|g', g'\rangle$. We have neglected those coupleings, assuming that resonance conditions for their survival in the interaction picture are not satisfied.

### IV. SPIN-SPIN INTERACTION MODELS AND TWO MODE APPROXIMATIONS

In this section, we will be primarily concerned with process which produce entanglement of atomic spin states. We therefore consider a spin 1/2 system and adopt the notation $|\uparrow\rangle$ for spin up and $|\downarrow\rangle$ for spin down, with the following operators and identities:

$$ \sigma_x = |\uparrow\rangle \langle \downarrow| - |\downarrow\rangle \langle \uparrow|, $$

$$ \sigma_+ = \frac{1}{2}(\sigma_x + i\sigma_y) = |\uparrow\rangle \langle \downarrow|, $$

$$ \sigma_- = \frac{1}{2}(\sigma_x - i\sigma_y) = |\downarrow\rangle \langle \uparrow|, $$

$$ \sigma_+^2 - \sigma_-^2 = 0, $$

$$ \sigma_+ \sigma_- + \sigma_- \sigma_+ = 1, $$

$$ \sigma_z^2 = \sigma_y^2 = 1. $$

The Bragg/Raman coupling as discussed here is of the form

$$ \frac{\hbar \Omega_{R}}{2} \left[ (|\uparrow\rangle \langle \downarrow|_1 \otimes (|\uparrow\rangle \langle \downarrow|)_2 + (|\downarrow\rangle \langle \uparrow|)_1 \otimes (|\downarrow\rangle \langle \uparrow|)_2 \right] $$

$$ = \frac{\hbar \Omega_{R}}{2} (\sigma_+^{(1)} \otimes \sigma_+^{(2)} + \sigma_-^{(1)} \otimes \sigma_-^{(2)}), $$

if we designate $|g\rangle$ and $|g'\rangle$ as $|\uparrow\rangle$ and $|\downarrow\rangle$, respectively. The Rabi frequency $\Omega_R$ is the two-photon Rabi frequency for transitions between $|\uparrow\rangle$ and $|\downarrow\rangle$ via the intermediate molecular state. Hence it contains the single photon Rabi frequencies $\Omega_1$ and $\Omega_2$, as well as the free-bound transition amplitude, $\Omega_{mb}$. This is a new type of coupling not widely discussed before. Its ability, however, to generate entanglement should be obvious as two atoms perform conditional evolution at all times, similar to the coupling for photon down conversion. Its prospects for creating massive GHZ states will now be studied.

We first compare our coupling scheme with other relevant models. In the original scheme of Sorensen and Mølmer [6], the two atom coupling takes the following form,

$$ \frac{\hbar \Omega_{R}}{2} \sigma_+^{(1)} \otimes \sigma_x^{(2)} $$

$$ = \frac{\hbar \Omega_{R}}{2} (|\uparrow\rangle \langle \downarrow| + |\downarrow\rangle \langle \uparrow|)_1 \otimes (|\uparrow\rangle \langle \downarrow| + |\downarrow\rangle \langle \uparrow|)_2, $$

a form different from ours. It is convenient to analyze such models for the case of many atoms in terms of collective spin operator $J_{z=x,y,z} = \frac{1}{2} \sum_{i} \sigma_{p}^{(i)}$. One can then show that the Hamiltonian for the Sorensen and Mølmer scheme becomes

$$ \sum_{i<j} \frac{\hbar \Omega_{R}}{2} \sigma_+^{(i)} \otimes \sigma_x^{(j)} \hbar \Omega_{R} \left( J_z^2 - \frac{N}{4} \right), $$

while our model gives

$$ \sum_{i<j} \frac{\hbar \Omega_{R}}{2} \left( \sigma_+^{(i)} \otimes \sigma_+^{(j)} + \sigma_-^{(i)} \otimes \sigma_-^{(j)} \right) $$

$$ = \frac{\hbar \Omega_{R}}{4} \left( J_x^2 + J_y^2 \right) $$

$$ = \frac{\hbar \Omega_{R}}{2} \left( J_x^2 - J_y^2 \right). $$

Recently, a many body, two mode coupling was proposed by Zoller and coworker [6]. They considered an interacting, two component (i.e. $|\downarrow\rangle$ and $|\uparrow\rangle$) condensate whose many body Hamiltonian becomes

$$ H = \sum_{j=\downarrow,\uparrow} \epsilon_{0,j} \int d\vec{r} |\phi_0(\vec{r})|^2 a_j^\dagger a_j $$
\[ + \frac{1}{2} \sum_{j = 1}^{N} U_{j} \int d\vec{r} |\phi_0(\vec{r})|^{4} a_j^\dagger a_j^\dagger a_j a_j + U_{i,j}^\dagger \int d\vec{r} |\phi_0(\vec{r})|^{4} a_i^\dagger a_i^\dagger a_j a_j. \] 

(39)

under the two mode approximation, where each component has the same spatial mode,

\[ \Psi_j(\vec{r}) = \phi_0(\vec{r})a_j. \] 

(40)

Introducing the collective spin operators

\[ J_x = \frac{1}{2} \left( a_0^\dagger a_1 + a_1^\dagger a_0 \right), \]
\[ J_y = \frac{i}{2} \left( a_0^\dagger a_1 - a_1^\dagger a_0 \right), \]
\[ J_z = \frac{1}{2} \left( a_1^\dagger a_1 - a_0^\dagger a_0 \right), \]

(41)

and taking the index 0 for \(| \downarrow \rangle\) and 1 for \(| \uparrow \rangle\) [13, 27, 28], one can show that \( J_{x,y,z} = \frac{1}{2} \sum_i \sigma_i^{x,y,z} \) as was used earlier. We also note that \( N = a_1^\dagger a_1 + a_0^\dagger a_0 \) and the Casimir relation

\[ J_{x}^2 + J_{y}^2 + J_{z}^2 = \frac{N}{2} \left( \frac{N}{2} + 1 \right). \] 

(42)

The interaction term Eq. (39) then becomes

\[ \frac{1}{2} J_{z}^2 (U_{00} + U_{11} - 2U_{01}) \int d\vec{r} |\phi_0(\vec{r})|^{4}. \] 

(43)

In the discussions of the numerical solutions to follow, we will use the second quantized notation, and consider a pure initial state with a fixed, total number of atoms \( N \). Specifically, we compare the three types of coupling,

\[ V_M = \hbar \Omega_R J_{z}^2, \] 

(44)

considered by Sorensen and Molmer [4], the spin squeezing model

\[ V_S = \hbar \Omega_R J_{z}^2, \] 

(45)

recently considered by Zoller and coworkers [7], and our proposed coupling

\[ V_{B/R} = \frac{\hbar \Omega_R}{2} (J_x^2 - J_y^2). \] 

(46)

Given an initial state \( |\psi(0)\rangle = a_1^\dagger N |\text{vac}\rangle / \sqrt{N} \), i.e. with all atoms condensed in state \(| \uparrow \rangle\), the time evolution operator for \( V_M \) is analytically known and takes a simple form at \( \Omega_R t = \pi/2 \) for \( N \) an even integer [4].

\[ U_M = e^{-iJ_{z}^2(\Omega_R t)} = e^{i\pi N/2 + i\pi/4} \frac{1}{\sqrt{N!}} a_0^\dagger a_1^\dagger a_1 N + e^{-i\pi/4}. \] 

(47)

It creates the maximally entangled \( N \)-GHZ state

\[ |\text{GHZ}_N\rangle \propto \frac{1}{\sqrt{2}} \left[ |0\rangle_0 |N\rangle_1 + i^{N+1} |N\rangle_0 |0\rangle_1 \right]. \] 

(48)

In the context of Bose-Einstein condensation, state \(|\text{GHZ}_N\rangle\) is an example of the interesting fragmented condensate [23]. The Sorensen and Molmer model (Eq. (44)) produces perfect GHZ-type states at selected times, while our coupling (Eq. (45)) doesn’t, in general, produce exact GHZ-type state. From numerical simulations, however, we find that our model, on average, produces more than 50% overlap with GHZ-type state (Eq. (45)) at selected times. Because of the geometric equivalence between \( J_{z}^2 \) and \( J_{x}^2 \), the spin squeezing model (Eq. (45)) also produces a perfect GHZ at selected times, except now the entanglement is between single particle states \(| \pm \rangle = (| \uparrow \rangle \pm | \downarrow \rangle)/\sqrt{2} \) and the initial state has to become \(| \pm \rangle \otimes |N\rangle \).

For our model, we were not able to find a general analytic form of the time evolution operator, even at specific values of \( t \). To numerically calculate the time evolution, we expand the wave function as

\[ |\psi(t)\rangle = \sum_{m=0}^{N} c_m(t) |N - m\rangle_0 |m\rangle_1, \] 

(49)

with the initial conditions given by the \( c_m(0) \)’s.

In Figs. 2 and 3 we show, for comparison, the generation of the maximum entangled GHZ state Eq. (48) as determined by model \( V_M \) (Eq. (44)) and our “molecular diffraction” model (Eq. (45)). We see that while the Sorensen and Molmer model creates perfect massive GHZ states at selected times, our model can also creates significant overlap with the massive GHZ at selected times.

We can also compare the achievable spin squeezing between our model and that of \( V_S \) (Eq. (45)), using the squeezing parameter

\[ \xi^2 = \frac{N(\Delta \bar{a}_{i}'s)^2}{\langle \bar{a}_{i}'s \rangle^2 + \langle \bar{a}_{i}'s \rangle^2}, \] 

(51)

where \( \bar{a}_i, i = 1, 2, 3 \) are mutually orthogonal unit vectors. Other discussions of spin squeezed states can be found in [18, 31, 32].

For the initial state

\[ |\psi(0)\rangle = |+\rangle^\otimes N = \frac{1}{2^{N/2}} \frac{1}{\sqrt{N!}} \left( a_0^\dagger + a_1^\dagger \right)^N |\text{vac}\rangle, \] 

(52)

it has been shown [8] that \( \xi^2(t > 0) < 1 \) for some set of \( \bar{a}_i \)’s. This model of \( V_S \) (Eq. (13)) is in fact the one-axis twisting model considered by Kitagawa and Ueda earlier [15]. In this case the problem can be solved analytically, with the result

\[ (\Delta \bar{a}_{i}'s)^2 \sim N^{1/3}. \] 

(53)

On the other hand, our model resembles the two-axis twisting model of Kitagawa and Ueda [13], and has to
be solved numerically. In the limit of large $N$ and with the condensate initially in $|\uparrow\rangle^\otimes N$, one can show that for an initial $J_z = N/2$ ($\vec{J}$-spin pointing along the positive $z$-direction),

$$ (\Delta J_{\hat{r}_1})_{\text{min}}^2 \sim \frac{1}{2}. \quad (54) $$

The optimal squeezing in this case occurs with $J_z + J_y$. This result can be easily verified by taking a semiclassical approximation in the dynamical equation for $J_x$ and $J_y$. We find that the time scale of reaching maximum squeezing is $\sim 1/(N\Omega_R)$ (see also \cite{33}). For condensates containing $10^6$ atoms, even with a very weak coupling $\Omega_R \sim 1$ (Hz), the maximum squeezing is reached within a micro-second.

In Figure 4 the squeezing parameters and the corresponding angle $\theta_{\text{min}}$ are computed numerically for the model $V_S$ (Eq. (45)) with the initial state given by Eq. (52). Fig. 5 is a similar calculation for our model (Eq. (46)) starting with all atoms in state $|\uparrow\rangle$. Our model achieves a better squeezing at an earlier time. In addition, the phase angle $\theta_{\text{min}}$ is fixed at the value $\pi/4$. More detailed discussions of the spin squeezing properties of our model can be performed, including dissipation and finite system size \cite{34,35,36}.

Finally, we also note that our coupling scheme for si-
multistate transitions to two different final states gives
\[
V_{B/R}^{[2]} = \frac{\hbar \Omega R}{4} \left[ (a_1^\dagger a_0^\dagger a_1 a_0 + a_0^\dagger a_0^\dagger a_1 a_1) + h.c. \right].
\] (55)
In the undepleted pump limit when dissipation exists, or when the coupling efficiency is small, this coupling can be approximated by
\[
V_{B/R}^{[2]} \approx \mathcal{E} \left( a_1^\dagger a_1^\dagger + a_1^\dagger a_0^\dagger \right).
\] (56)
This is similar to the recent ideas of Refs. [8, 9, 19] which creates entangled EPR pairs every time an event occurs, just like in a parametric down conversion process [37, 38].

V. DETECTION OF THE MAXIMALLY ENTANGLED STATES

While it seems that there are promising schemes to generating massive entanglement of condensate atoms, the detection of such entanglement represents a significant challenge by itself as it is very difficult to perform individual atom resolved measurements [32, 33] in a condensate. Recently, it was suggested that performing a time reversed dynamic evolution can detect the coherence of the created N-GHZ state [32, 33].

In this section, we detail the working mechanism of the proposed scheme by performing explicit calculations. We will use the $V_M$ model as an example. To simplify the algebra, we will use $\tau = \Omega R t$, and denote the basis states according to
\[
|n_1\rangle \equiv |n_0, n_1\rangle = |n_0\rangle |n_1\rangle_1.
\] (57)
Assume we start with $|\uparrow\rangle^\otimes N$, then at $\tau = \pi/2$, a maximally entangled state
\[
|\psi(\tau = \pi/2)\rangle \propto \frac{1}{\sqrt{2^N}}(|N\rangle + i^{N+1}|0\rangle),
\] (58)
Immediately reversing the time dynamics ($V_M \to -V_M$), it is easy to show that after another $\tau = \pi/2$, we recover the initial state $|\uparrow\rangle^\otimes N$, i.e. all atoms to be detected are in state $|\uparrow\rangle$. If instead, the created state at $\tau = \pi/2$ is a completely incoherent mixture of $|0\rangle$ and $|N\rangle$, given by the (N-body) density matrix
\[
\rho(\tau = \pi/2) = \frac{1}{2} |0\rangle \langle 0| + \frac{1}{2} |N\rangle \langle N|,
\] (59)
then the time reversed dynamics will lead to observations of variable number of atoms in state $|\downarrow\rangle$ as well.

For the general case, let’s assume the restricted form
\[
\rho(\tau = \pi/2) = p_0 |0\rangle \langle 0| + p_N |N\rangle \langle N| + \beta |0\rangle \langle N| + \beta^* |N\rangle \langle 0|,
\] (60)
with $0 \leq p_0/N \leq 1$, $p_0 + p_N = 1$, and $|\beta|^2 \leq p_0 p_N$. Time reversed evolution of $V_M$ for $\tau$ from $\pi/2$ will lead to
\[
\rho(\tau + \pi/2) = U_M(-\tau) \rho(\pi/2) U_M(-\tau) = e^{i J_\tau^Z} \rho(\pi/2) e^{-i J_\tau^Z},
\] (61)
which can be explicitly evaluated with the use of rotation operation
\[
e^{\pm i J_\tau^Z} = e^{-i \frac{\pi}{2} J_y} e^{\pm i J_\tau^Z} e^{i \frac{\pi}{2} J_y},
\] (62)
and the disentangling relation
\[
e^{-i \frac{\pi}{2} J_y} = e^{J_x + J_y \ln 2} e^{-J_x} = e^{-J_x - J_y \ln 2} e^{J_x},
\] (63)
e^{i \frac{\pi}{2} J_y} = e^{-J_x + J_y \ln 2} e^{-J_x} = e^{-J_x - J_y \ln 2} e^{J_x}. (64)

Denote
\[
A = \langle N | e^{J_x + J_y \ln 2} e^{-J_x} | 0 \rangle,
\] (65)
\[
B = \langle N | e^{J_x + J_y \ln 2} e^{-J_x} | N \rangle,
\] (66)
then we find
\[
\rho_{NN}(\tau + \pi/2) = \langle N | \rho(\tau + \pi/2) | N \rangle
\] (67)
with
\[
A = \sum_{m=0}^{N} \frac{N!}{m!(N-m)!} (-1)^m e^{i(m-\frac{\beta}{2})^2 \tau},
\] (68)
\[
B = \sum_{m=0}^{N} \frac{N!}{m!(N-m)!} e^{i(m+\frac{\beta}{2})^2 \tau}.
\] (69)
The following angular momentum algebra has been used,
\[
J_+ |m\rangle = \sqrt{(m+1)(N-m)} |m+1\rangle,
\] (70)
\[
J_- |m\rangle = \sqrt{(m(N-m+1)} |m-1\rangle,
\] (71)
\[
J_z |m\rangle = (m - \frac{N}{2}) |m\rangle,
\] (72)
\[
\langle m | J_+ = (J_- | m \rangle)^\dagger.
\] (73)
At $\tau = \frac{\pi}{2}$, we find
\[
\rho_{NN}(\pi) = \frac{1}{2} + (-1)^{N} \text{Im} (\beta) \leq 1,
\] (74)
because $|\text{Im} (\beta)| \leq |\beta| \leq \sqrt{p_0 p_N} \leq 1/2$, for any state. Thus we conclude that the probability of detecting atoms in state $|\downarrow\rangle$ is nonzero for mixed states. In fact, for the completely incoherent mixture Eq. (63) with $p_0 = p_N = \frac{1}{2}$ and $\beta = 0$, $\rho_{00}(\pi) = \rho_{NN}(\pi) = 1/2$.

It’s interesting to compare the average populations and fluctuations of spin up $|\uparrow\rangle$ number operator $n_1 = a_1^\dagger a_1$ after the time reversed evolution for $\tau = \frac{\pi}{2}$ starting with the following different initial conditions,

- A pure state,
\[
|\psi(\pi/2)\rangle = \frac{1}{\sqrt{2^N}}(|N\rangle + i^{N+1}|0\rangle)
\] (75)
\[
\langle n_1(\pi) \rangle = \text{tr}(\rho n_1) = N,
\] (76)
\[
\langle n_1^2(\pi) \rangle = \text{tr}(\rho n_1^2) = N^2,
\] (77)
\[
\Delta n_1(\pi) = \sqrt{\langle n_1^2 \rangle - \langle n_1 \rangle^2} = 0;
\] (78)
Thus, these two cases become easily distinguishable. In Fig. 6, we compare the time dependent results of averaged population in state $|↑\rangle$ and its variance $\Delta n_1$ for spin up $|↑\rangle$ component. The solid lines are for starting in the N-GHZ state Eq. (58) while the dashed lines are for starting in the incoherent mixture Eq. (59). The upper two lines are for $\langle n_1 \rangle$ while the lower ones are for $\Delta n_1$.

- An incoherent mixture, 
  \[ \rho(\pi/2) = \frac{1}{2}(|N\rangle \langle N| + |0\rangle \langle 0|) \]
  \[ \langle n_1(\pi) \rangle = \text{tr}(\rho n_1) = \frac{N}{2}, \]
  \[ \langle n_1^2(\pi) \rangle = \text{tr}(\rho n_1^2) = \frac{N^2}{2}, \]
  \[ \Delta n_1(\pi) = \frac{N}{2}. \]  

Thus, these two cases become easily distinguishable. In Fig. 6, we compare the time dependent results of averaged population in state $|↑\rangle$ and its variance, from the time reversed evolution gives identical results that demonstrate that the time reversed dynamics is in fact independent of $\tau$. We find that the width of the Gaussian like features near $\Omega_R t \sim \pi$ is inversely proportional to $\sqrt{N}$, but the plateau for the variance $\Delta n_1$ is at around $N/\sqrt{3}$.

For factorizable N-atom states, whether a pure state $|\psi(\pi/2)\rangle = \frac{1}{2^{N/2}}(|↑\rangle + |↓\rangle)^{\otimes N}$ or a mixture $\rho(\pi/2) = \frac{1}{2^{N/2}}(|↑\rangle \langle ↓| + |↓\rangle \langle ↑|)^{\otimes N}$, the probability distribution after the time reversed evolution gives identical results that are different from the two multi-atom correlated states discussed earlier. In fact, the final results simply reflects the binomial distributions of $n_1$ in a equal superposition separable state and is given by

\[ \langle n_1(\pi) \rangle = \text{tr}(\rho n_1) = \frac{N}{2}, \]
\[ \langle n_1^2(\pi) \rangle = \text{tr}(\rho n_1^2) = \frac{N(N+1)}{4}, \]

\[
\Delta n_1(\pi) = \frac{\sqrt{N}}{2}.
\]  

In this particular example with $V_M$ as the interaction, the above results actually is independent of $\tau$. Thus the measurement of total single particle state population fluctuations after the time reversed evolution provides a strong indication for the coherence and correlation of the created entangled states.

\section{VI. LIMITATIONS AND ADVANTAGES}

There are several limitations to our model that needs to be discussed carefully. First, the molecular coupling needs to dominate over the atomic coupling; i.e., we need to achieve a molecular coupling

\[ \Omega_R^M = \frac{\Omega_1 \Omega_2^*}{\Delta_M} \eta_{b\sigma} \eta_{b'\sigma'}^* \]
\[
\gg \Omega_R^A = \frac{\Omega_1 \Omega_2^*}{\Delta_A},
\]

where $\Delta_{M/A}$ are detunings from the molecular and atomic intermediate states, respectively. We can estimate analytically the free-bound transition amplitude using the interacting ground state wave function from [25], and excited state wave function from the bound state solutions in $\pi - C_3/r^3$ potential modified by an inner repulsive core [40]. Most likely, however, the deepest molecular bound state with significant amplitude should be chosen in order to maximize the detuning from the atomic transition. In addition, we should have $\Delta_M \gg \gamma_M$ to minimize spontaneous emission which would lead to decoherence and loss of atoms.

A clear advantage of our scheme is the signature for diffraction via a molecular intermediate state. The diffracted atoms would move at half the speed of atoms that have undergone atomic Bragg diffraction. Even as the number of atoms is increased, our scheme achieves the same level of squeezing and the same high value of overlap with the massive GHZ state.

Our scheme is based on an engineered interaction that can be turned on and off like the scheme of Sørensen and Mølmer (Eq. (14)). In particular, our scheme works for non-interacting atoms which can decrease the noise due to atom-atom interactions in a $U(1)$ symmetry breaking condensate state. We have also provided explicit calculations that demonstrate that the time reversed dynamics can be used to verify the coherence of the intended maximally entangled states.

Among all models discussed, there exists an even/odd number problem: The form of the final state depends on whether the total number of condensed atoms is even or odd number. If, however, the condensate resembles a coherent superposition of number states (grand canonical ensembles), we should average the results over the number distribution $p(N)$ as well. This will be explored in a future study.
This work benefited from our participation of the 2000 workshop on quantum degenerate gases at the Lorentz Center, University of Leiden. We thank H. Stoof for the hospitality. We thank Dr. M. S. Chapman and S. L. Rolston for helpful discussions. This work is supported by a grant from NSA, ARDA, and DARPA under ARO Contract No. DAAD19-01-1-0667, and by a grant from the NSF PHY-0113831.

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