Conversion Efficiency of Ultracold Fermionic Atoms to Bosonic Molecules via Feshbach Resonance Sweep Experiments

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We explain why the experimental efficiency observed in the conversion of ultracold Fermi gases of ^{40}K and ^{6}Li atoms into diatomic Bose gases is limited to 0.5 when the Feshbach resonance sweep rate is sufficiently slow to pass adiabatically through the Landau-Zener transition but faster than "the collision rate" in the gas, and increases beyond 0.5 when it is slower. The 0.5 efficiency limit is due to the preparation of a statistical mixture of two spin states, required to enable *s*-wave scattering. By constructing the many-body state of the system we show that this preparation yields a mixture of even and odd parity pair states, where only even parity can produce molecules. The odd parity spin-symmetric states must decorrelate before the constituent atoms can further Feshbach scatter, thereby increasing the conversion efficiency; "the collision rate" is the pair decorrelation rate.

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The formation of quantum-degenerate molecular gases by association of quantum-degenerate atomic gases has been the subject of intense theoretical deliberation in recent years. Experimental efforts soon followed, demonstrating the association of an atomic Bose-Einstein condensate into ground-state molecules either via a stimulated Raman transition or through a magnetic-field controlled Feshbach resonance [1,2].

In four recent experiments a Fermi gas of atoms was converted into an ultracold Bose gas of molecules by adiabatic passage through a Feshbach resonance [3–6]. In two of these experiments [3,6] ultracold $^{40}K_2$ molecules were produced from a quantum-degenerate Fermi gas of ^{40}K atoms, whereas in Refs. [4,5] ^{6}Li atoms were converted to diatomic molecules. An interesting feature of these studies is that when the sweep rate through the Feshbach resonance was comparable to the background elastic-scattering rate, yet slow with respect to the atom-molecule coupling rate, a maximum atom-molecule transfer efficiency of 50% was reported [3,4]. However, when the sweep rate was much slower, approaching "close to thermal equilibrium" conditions, higher conversion efficiency was attained [5,6].

In this Letter we show that the observed 50% saturation of the atom-molecule conversion efficiency in the fast (yet adiabatic) Feshbach sweep regime is a result of the initial state preparation. The fermionic atoms in the experiments of Refs. [3–6] are prepared in statistical mixtures of two spin states in order to enable their *s*-wave scattering. Elastic-scattering collisions between atoms in these different spin states results in cooling of the trapped gas as hot atoms leave the trap. This procedure effectively splits the many-body system into two subsystems corresponding to two different spin states, as shown in Fig. 1. If we consider pairs of atoms, one from each spin state (where spin means total spin— $|\tilde{f}m_f\rangle$ —not electronic spin [7]), half the pairs are antisymmetric spin states (for ^{6}Li these are spin singlets) and half are symmetric spin states (for ^{6}Li, spin triplets). More explicitly, the reduced two-particle density matrix obtained by tracing out all but one particle of one spin state and another particle of the other spin state contains 50% spin antisymmetric (spin singlet for ^{6}Li), interacting via an *s* wave (even parity spatial state), and 50% spin symmetric (e.g., the triplet spin-symmetric superposition of the form $|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$ for ^{6}Li), interacting via a *p* wave (odd- parity spatial state). Sweeping through the resonance sufficiently slowly so the Landau-Zener transition [8] is traversed adiabatically [1], spin-singlet pairs are converted to molecules.

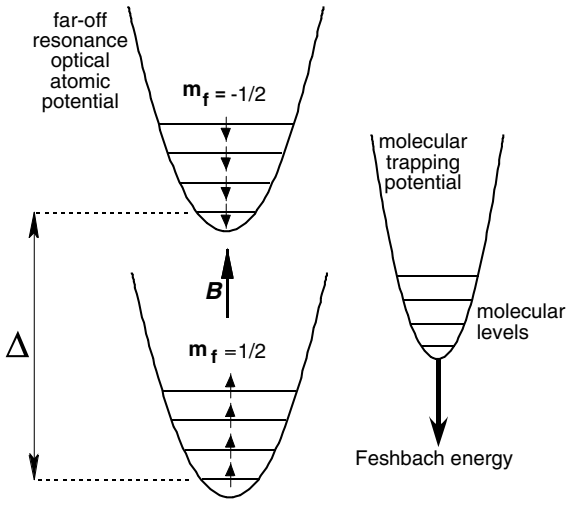

FIG. 1. Schematic drawing of the trapping potentials for atoms with different spin projections, m_a and m_b (explicitly labeled ↑ and ↓ as appropriate for the ^{6}Li case), and for diatomic molecules (on the right side of the figure) whose potential is swept downward in energy as a function of time. Δ is the Zeeman energy splitting. The molecular potential force constant is twice as large as that of the atoms, but the energy level spacings of the atoms and molecules are equal.

However, since the Feshbach is an s-wave resonance, spin-triplet pairs cannot Feshbach scatter. Once a triplet state is formed, its constituent atoms can undergo triplet/p-wave collisions with other atoms only until they are decorrelated. Hence, if collisional dephasing is slow with respect to the sweep rate, colliding pairs of atoms form either spin singlets (eventually producing a molecule via the Feshbach resonance sweep) or spin triplets (producing a correlated pair that cannot Feshbach scatter). If the sweep rate is slower than the time required for spin-triplet states to decorrelate, the conversion efficiency can grow beyond 50%.

Both the ^{6}Li and ^{40}K Feshbach experiments involve two atomic spin states $|\chi\rangle \equiv |\tilde{f}_\chi m_\chi\rangle$ with $\chi = \{a, b\}$. An external magnetic field generates a large Zeeman splitting Δ between the spin- states $|a\rangle$, $|b\rangle$, as depicted in Fig. 1. We consider the Feshbach atom-molecule coupling Hamiltonian

$$\hat{H}_F = g\sum_{\mathbf{q},\mathbf{k}} \hat{a}^\dagger_{\mathbf{q}/2-\mathbf{k}} \hat{b}^\dagger_{\mathbf{q}/2+\mathbf{k}} \hat{c}_\mathbf{q} + \text{H.c.}, \tag{1}$$

where g is the coupling coefficient, $\hat{a}^\dagger_{\mathbf{q}/2-\mathbf{k}}$ and $\hat{b}^\dagger_{\mathbf{q}/2+\mathbf{k}}$ are the usual atomic creation operators for spin states $|a\rangle$ and $|b\rangle$, respectively, and $\hat{c}_\mathbf{q}$ is the molecular annihilation operator. Written explicitly for fermionic operators $\hat{a}_\mathbf{k}$, $\hat{b}_\mathbf{k}$, and bosonic $\hat{c}_\mathbf{q}$, the Hamiltonian (1) takes the form

$$\hat{H}_F = \frac{g}{2}\sum_{\mathbf{q},\mathbf{k}} (\hat{a}^\dagger_{\mathbf{q}/2-\mathbf{k}} \hat{b}^\dagger_{\mathbf{q}/2+\mathbf{k}} - \hat{b}^\dagger_{\mathbf{q}/2-\mathbf{k}} \hat{a}^\dagger_{\mathbf{q}/2+\mathbf{k}}) \hat{c}_\mathbf{q} + \text{H.c.}, \tag{2}$$

having used the anticommutation relation $\{\hat{a}^\dagger_\mathbf{k}, \hat{b}_{\mathbf{k}'}\} = 0$.

It is evident that the fermionic Feshbach coupling Hamiltonian (2) is antisymmetric under spin permutation and that it has even parity. It only allows for the association of atom pairs colliding with even parity partial waves (effectively restricted at ultracold temperatures, to s-wave scattering) with an antisymmetric spin state (a spin singlet in the ^{6}Li case). Defining the commuting operators

$$\hat{S}_\mathbf{q} = \sum_\mathbf{k} (\hat{a}_{\mathbf{q}/2-\mathbf{k}} \hat{b}_{\mathbf{q}/2+\mathbf{k}} - \hat{b}_{\mathbf{q}/2-\mathbf{k}} \hat{a}_{\mathbf{q}/2+\mathbf{k}}), \tag{3}$$

$$\hat{T}_\mathbf{q} = \sum_\mathbf{k} (\hat{a}_{\mathbf{q}/2-\mathbf{k}} \hat{b}_{\mathbf{q}/2+\mathbf{k}} + \hat{b}_{\mathbf{q}/2-\mathbf{k}} \hat{a}_{\mathbf{q}/2+\mathbf{k}}), \tag{4}$$

the Feshbach coupling Hamiltonian for fermionic atoms simply reads

$$\hat{H}_F = \frac{g}{2}\sum_\mathbf{q} \hat{S}^\dagger_\mathbf{q} \hat{c}_\mathbf{q} + \text{H.c.} \tag{5}$$

We note parenthetically that for bosonic operators $\hat{a}_\mathbf{k}$, $\hat{b}_\mathbf{k}$, Eq. (1) assumes the form $\hat{H}_F = \frac{g}{2}\sum_\mathbf{q} \hat{T}^\dagger_\mathbf{q} \hat{c}_\mathbf{q} + \text{H.c.}$

As discussed in Refs. [3,4], if all the fermions are in a pure superposition of internal spin states, e.g., $\alpha|a\rangle + \beta|b\rangle$, the colliding atoms will have a symmetric spin state and therefore any pair is allowed to interact only through odd parity partial waves (e.g., p wave). In order to enable the s-wave scattering of atoms, a statistical mixture of two Fermi gases corresponding to two spin states is obtained by rapidly driving the Zeeman transition in the presence of a magnetic-field gradient. As different atoms are driven at different frequencies, the above process results in an inhomogeneous broadening of the transition. Since all coherence between the two spin states is thus destroyed, the initial state of the system, after cooling, is best described by a density matrix composed of an *incoherent mixture* of two decoupled Fermi gases, one for each spin state:

$$\rho_0^N = \mathcal{A}\{[\rho_{N/2}(a, T)|a\rangle^{(N/2)(N/2)}\langle a|] \otimes (\rho_{N/2}(b, T)|b\rangle^{(N/2)(N/2)}\langle b|)\}\mathcal{A}^{-1}, \tag{6}$$

where $\rho_{N/2}(\chi, T)$ denotes the motional part of the density matrix for $N/2$ atoms with spin projection $\chi = \{a, b\}$ at temperature T, and $\mathcal{A}$ is the antisymmetrization operator of all the particles.

Before the Feshbach sweep begins, cooling of the two Fermi gases induces pairwise and many-body correlations. As opposed to a thermal gas, the probability for forming a molecule in a single Feshbach collision is determined by the symmetry of the many-body state. It is instructive to explicitly construct the many-body state of the cooled system. To do so we start by antisymmetrizing each of the wave functions for the two Fermi seas separately. Since atoms in each Fermi sea have the same spin projection, their many-body wave function is a tensor product of an odd parity Slater determinant for the spatial degrees of freedom and a totally symmetric spin configuration. It is evident from the construction of the pertinent Young tableaux (see Fig. 2) that the antisymmetrization of the *two* Fermi seas can be done in only two ways: a totally antisymmetric spatial wave function times a spin-symmetric configuration $|F = N/2, M_F = 0\rangle$ (the row Young tableau on the right hand side of the equation in Fig. 2), and a state which is spin antisymmetric upon the exchange of particles of different spin corresponding to $|F = 0, M_F = 0\rangle$ (the second tableau on the right hand

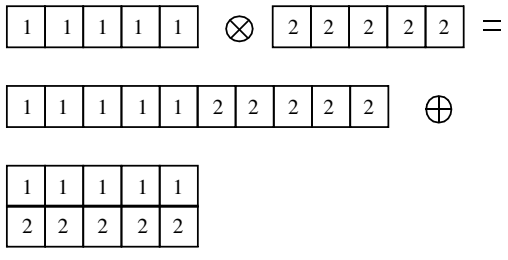

FIG. 2. Spin degrees of freedom Young tableaux corresponding to a totally spin-symmetric configuration for each of the Fermi seas (each row tableau corresponds to a different spin projection m_a denoted by 1 and m_b denoted by 2) and its decomposition into a totally symmetric $F = N/2$ and a $F = 0$ many-body configuration. Young tableaux for spatial degrees of freedom are obtained by interchanging rows and columns.

side of the equation in Fig. 2). This is the many-body analogy of the two spin 1/2 cases in which the two-body state naturally falls into two classes: symmetric or antisymmetric in the spatial coordinates corresponding to spin-antisymmetric (singlet) and spin-symmetric (triplet) states, respectively, such that the total fermionic wave function is antisymmetric with respect to the exchange of the two particles.

Let us consider the reduced two-particle density matrix $\rho_0^{(2)} = \mathrm{Tr}_{(N-2)}(\rho_0^N)$ obtained by tracing out all but one particle with spin projection a and another particle with spin projection b. It is readily seen from the Young tableaux in Fig. 2 that tracing out all but two of the particles, one with spin up and the other with spin down, leaves a two-particle reduced density matrix which is an equally weighted sum of a spin-symmetric (two box row tableau) and a spin-antisymmetric (two box column tableau) atomic pair. Therefore, if a given atom pair is spatially antisymmetric, then each of the atoms comprising this pair will be spatially antisymmetric with any other atom of opposite spin due to the symmetry of the many-body state. This antisymmetrization induces atomic correlations and therefore determines collision probabilities. The spin part of $\rho_0^{(2)}$ is identical for all atomic pair states, and is obtained by projection onto the momentum states of any given colliding pair,

$$\langle \mathbf{q}/2 - \mathbf{k}|\langle \mathbf{q}/2 + \mathbf{k}|\rho_0^{(2)}|\mathbf{q}/2 + \mathbf{k}\rangle|\mathbf{q}/2 - \mathbf{k}\rangle = \tfrac{1}{2}(|a\rangle|b\rangle\langle b|\langle a| + |b\rangle|a\rangle\langle a|\langle b|) = \langle \mathbf{q}|\tfrac{1}{2}(|S\rangle\langle S| + |T\rangle\langle T|)|\mathbf{q}\rangle. \quad (7)$$

where $|\mathbf{q}/2 \pm \mathbf{k}\rangle$ is the single-particle spatial wave function with momentum $\mathbf{q}/2 \pm \mathbf{k}$, $|T\rangle = (|a\rangle|b\rangle + |b\rangle|a\rangle)/\sqrt{2}$ is a spin-symmetric triplet state, and $|S\rangle = (|a\rangle|b\rangle - |b\rangle|a\rangle)/\sqrt{2}$ is a spin-antisymmetric (singlet) state. In the middle line of Eq. (7) we have omitted the projection onto momentum states to simplify the notation. Equation (7) demonstrates the simple result that in the absence of coherence between colliding particles in different spin states, collisions with a spin triplet (via a p wave) are just as probable as collisions with a spin singlet (via an s wave). In the above expressions, an explicit orbital-spin form of the triplet state is given by

$$|T\rangle = [|\phi_a(1)\phi_b(2)\rangle - |\phi_b(1)\phi_a(2)\rangle][|f_a m_a, f_b m_b\rangle + |f_b m_b, f_a m_a\rangle], \quad (8)$$

where $\phi_a(i)$ is the orbital for the ith atom of species a. For example, for the ^{6}Li case,

$$|T\rangle = [|\phi_a(1)\phi_b(2)\rangle - |\phi_b(1)\phi_a(2)\rangle][|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle]. \quad (9)$$

This is similar for $|S\rangle$, but with the signs reversed.

We can describe the process of adiabatically scanning the magnetic field from high to low field in terms of a Landau-Zener transition between a pair of free atoms, one from each Fermi gas, into the resonant vibrational level of the diatomic molecule. Since only the spin-antisymmetric, even parity, pairs are coupled to molecules by the Feshbach Hamiltonian (5), the maximal anticipated conversion efficiency is 0.5. The spin-symmetric pairs cannot interact via the Feshbach resonance and therefore cannot be converted to molecules.

As long as pair correlations are maintained, constituent atoms of the spin-symmetric pairs remaining after Feshbach coupling cannot interact with another atom to produce a spin-antisymmetric state via elastic-scattering collisions. This can be demonstrated by considering the state formed by the tensor product of a spin-symmetric pair with another fermion and antisymmetrizing:

$$\begin{aligned}\mathcal{A}\{&[|\mathbf{k}\rangle_1|\mathbf{k}'\rangle_2 - |\mathbf{k}'\rangle_1|\mathbf{k}\rangle_2](|a\rangle_1|b\rangle_2 + |b\rangle_1|a\rangle_2) \otimes |\mathbf{k}''\rangle_3|a\rangle_3\} \\ &= [|\mathbf{k}\rangle_1|\mathbf{k}'\rangle_2 - |\mathbf{k}'\rangle_1|\mathbf{k}\rangle_2](|a\rangle_1|b\rangle_2 + |b\rangle_1|a\rangle_2) \otimes |\mathbf{k}''\rangle_3|a\rangle_3 \\ &\quad -[|\mathbf{k}\rangle_3|\mathbf{k}'\rangle_2 - |\mathbf{k}'\rangle_3|\mathbf{k}\rangle_2](|a\rangle_3|b\rangle_2 + |b\rangle_3|a\rangle_2) \otimes |\mathbf{k}''\rangle_1|a\rangle_1 \\ &\quad -[|\mathbf{k}\rangle_1|\mathbf{k}'\rangle_3 - |\mathbf{k}'\rangle_1|\mathbf{k}\rangle_3](|a\rangle_1|b\rangle_3 + |b\rangle_1|a\rangle_3) \otimes |\mathbf{k}''\rangle_2|a\rangle_2. \quad (10)\end{aligned}$$

Tracing out one of the particles results in only spin-symmetric pair states, implying that no spin-antisymmetric collisions are allowed until the spin-symmetric pair decoheres. Furthermore, using similar argumentation, it is also evident that pairs of spin-symmetric pairs cannot interact in such a way that two of the four atoms scatter as a spin-antisymmetric, even parity, pair. Equation (10) is just a specific three particle example of the general case, which is apparent by considering the Young tableaux in Fig. 2 from which one can infer that all two-particle states are either odd parity spin- ymmetric for all atoms, or even parity spin antisymmetric for all atoms with different spin projections. It explicitly depicts the case when one has to combine a two box row tableau with a further box tableau, and since the states with the same spin projection need to be symmetric this can be done only in one way. Therefore, until the spin-symmetric states decohere, allowing a finite probability for spin-antisymmetric (even parity) collisions, no further molecules can be made from their constituent atoms.

Slowing the sweep rate through the Feshbach resonance, $\gamma_f = \dot{B}(\Delta B)^{-1}$, where $B(t)$ is the time-dependent magnetic field and ΔB is the Feshbach resonance width, to the point where it is smaller than the pair decorrelation rate γ_{st} for triplet to singlet transitions, allows the two-particle spin-symmetric pairs not swept by the Feshbach

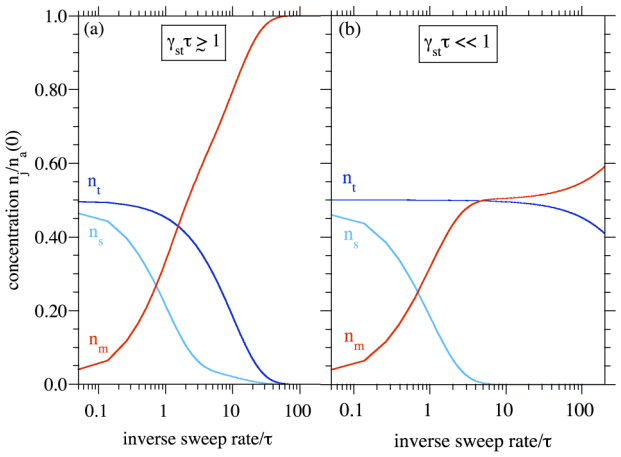

FIG. 3 (color online). Concentrations of molecules n_m, singletlike atomic pairs n_s, and tripletlike atomic pairs n_t versus inverse sweep rate for two limiting values of the pair decorrelation rate: (a) $\gamma_{st}\tau \geq 1$, (b) $\gamma_{st}\tau \ll 1$.

resonance to be destroyed by collisions. Consequently, further molecule-producing s-wave/spin-antisymmetric collisions can take place and higher than 50% efficiency may be obtained as reported by Cubizolles *et al.* [5] and Greiner *et al.* [6].

In Fig. 3 we schematically illustrate the expected final populations of singlet and triplet pairs, and molecules as a function of the Feshbach sweep rate. When the sweep rate γ_f is fast compared to the elastic-scattering rate in the gas τ^{-1}, the conversion efficiency is limited to less than 50% due to the Landau-Zener nature of the process [1,8] and the fact that only the spin-singlet pairs can participate. When the sweep rate is slow compared to τ^{-1}, the conversion efficiency depends on whether the time spent sweeping across the resonance is fast or slow compared to the pair decorrelation time γ_{st}. If $\gamma_f \gg \gamma_{st}$ only the spatially symmetric singlet portion is converted, leaving the spatially antisymmetric triplets unperturbed, and one is limited to 50% conversion. If γ_f is slow compared to γ_{st}, then as one sweeps, triplet and singlet states interconvert and one has the possibility of converting all the atoms. Consequently, we distinguish between two cases. When $\gamma_{st}\tau \geq 1$, i.e., the decorrelation process is fast compared to the Feshbach sweep rate, the conversion efficiency ranges from zero to unity, increasing with decreasing sweep rate as shown in Fig. 3(a). The adiabaticity criterion for achieving unit conversion efficiency is in this case $\gamma_f\tau \gg 1$. On the other hand, when the decoherence rate is slow compared to the sweep rate, $\gamma_{st}\tau \ll 1$, the new time scale γ_{st} plays a crucial role in determining conversion efficiency, as depicted in Fig. 3(b). For fast sweep rates, $\gamma_f\tau \gg 1$, adiabaticity is violated and the conversion efficiency is low. For slower sweeps such that $\gamma_{st}\tau \ll \gamma_f\tau \ll 1$, two-body adiabaticity is attained, but since the sweep is still too fast to allow triplet-singlet conversion the conversion efficiency stagnates at a broad 50% plateau as in the experiments of Refs. [3,4]. Only when the sweep rate is the slowest time scale in the system, $\gamma_f\tau \ll \gamma_{st}\tau \ll 1$, can the conversion efficiency limit to 100%, as expected from a truly many-body adiabatic sweep (over 50% observed in [5,6]).

In summary, we have explained the 0.5 limited atom to molecule transfer efficiency observed in experiments for fermionic atoms involving an adiabatic sweep through a Feshbach resonance. By constructing the many-body state we demonstrate that half the atoms are in a spatially antisymmetric state and therefore do not collide, whereas the other half are in a spin-symmetric state in which their collision can lead to the formation of a molecule. If the sweep is slow enough, half the atoms will convert to molecules, and the remaining atoms will not interact since any pair will be in a spatially antisymmetric state. We note that even though all remaining pairs are in a tripletlike correlated state, spontaneous transverse magnetization does not develop since the total m_f of any pair is zero, and this is a good quantum number. Significant modification of the collective excitation spectrum [9] and the free expansion profile of the remaining atomic Fermi gas are anticipated due to the complete lack of interactions, particularly in the vicinity of the Feshbach resonance. We further note that the remaining noninteracting Fermi gas of atoms can be made to interact by repeating the initial mixing procedure. Measurement of the conversion efficiency as a function of sweep rate in the whole range from very fast to very slow should serve to conclusively verify or contradict the picture of the conversion efficiency painted here.

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[1] F. H. Mies, E. Tiesinga, and P. S. Julienne, Phys. Rev. A **61**, 022721 (2000).
[2] E. A. Donley *et al.*, Nature (London) **417**, 529 (2002).
[3] C. A. Regal, C. Ticknor, J. L. Bohn, and D. S. Jin, Nature (London) **424**, 47 (2003).
[4] K. E. Strecker, G. B. Partridge, and R. G. Hulet, Phys. Rev. Lett. **91**, 080406 (2003).
[5] J. Cubizolles *et al.*, Phys. Rev. Lett. **91**, 240401 (2003).
[6] M. Greiner, C. A. Regal, and D. S. Jin, Nature (London) **426**, 537 (2003).
[7] The atomic state in a strong Zeeman field is $|\tilde{f}m_f\rangle = |\tilde{f}m_f(f, j, i, m_j, m_i)\rangle$ where m_f is a good quantum number, but f is not; $\tilde{f}$ is a quantum number that specifies a particular diagonalized state in the Paschen-Back limit.
[8] L. D. Landau, Z. Phys. Sowjetunion **2**, 46 (1932); C. Zener, Proc. R. Soc. London A **137**, 696 (1932).
[9] L. Vichi and S. Stringari, Phys. Rev. A **60**, 4734 (1999).