Formation of a molecular Bose-Einstein condensate and an entangled atomic gas by Feshbach resonance

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Processes of association in an atomic Bose-Einstein condensate, and dissociation of the resulting molecular condensate, due to Feshbach resonance in a time-dependent magnetic field, are analyzed incorporating non-mean-field quantum corrections and inelastic collisions. Calculations for the Na atomic condensate demonstrate that there exist optimal conditions under which about 80% of the atomic population can be converted to a relatively long-lived molecular condensate (with lifetimes of 10 ms and more). Entangled atoms in two-mode squeezed states (with noise reduction of about 30 dB) may also be formed by molecular dissociation. A gas of atoms in squeezed or entangled states can have applications in quantum computing, communications, and measurements.

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Introduction.— The recently discovered Bose-Einstein condensates (BEC), or matter waves, resemble in certain ways coherent electromagnetic radiation. This similarity stimulated the development of atomic optics [1], involving non-classical states of the atomic fields, such as squeezed and entangled states [2]. Squeezed states are characterized by noise reduction, and can be applied in communications and measurements. Entangled states of a decomposable system cannot be expressed as a product of the component states, and can be used in quantum computing and communications. Squeezed atomic states can be formed in four-wave mixing [3], in arrays of atomic traps [4], in multimode condensates [5, 6], in the decay of unstable BEC [7], in collisions of BEC wavepackets [8], and as the outcome of Bogolubov fluctuations subject to stimulated light scattering [9] or Belyaev dumping [10]. The squeezing can be measured experimentally by using homodyne detection, analogous to the one used in quantum optics (see Ref. [11]). The key component of this method — a beam splitter — already exists (see Ref. [12]).

The present work suggests the dissociation of molecular BEC as a source of atom fields in two-mode squeezed states that are entangled. Formation of single-mode squeezed states by the same mechanism has been discussed in Refs. [13, 14]. Formation of entangled atomic pairs in the dissociation of individual diatomic molecules has been considered in Ref. [15]. Other mechanisms of formation of entangled gases have been discussed in Refs. [16, 17].

The molecular BEC required as the source of the entangled gas is interesting in its own right, although it has not been realized yet. Formation of a molecular BEC by direct cooling of molecular gases is obstructed by the rotational degrees of freedom. An alternative method is the association of atomic BEC [15, 16]. A process of photoassociation [17], realized experimentally [18], is obstructed by spontaneous emission [19]. We consider here the association of atoms in BEC by Feshbach resonance [20] in a time-dependent magnetic field. Such a process is associated with a large condensate loss observed in recent experiments [19]. This loss follows from deactivation of the resonant molecules by inelastic collisions [16, 20], as well as from the formation of non-condensate atoms by molecular dissociation. An advantage of the use of Feshbach association is the possibility of reducing the negative effect of collisions by lowering the condensate density. Although the molecular BEC is made of excited molecules, and is therefore unstable, it still can be used as a source of atoms in entangled and squeezed states produced by the dissociation.

Reference [21] treats the condensate loss as a dissociation of single molecules. Many-body effects have been incorporated in Ref. [22] by introducing a width to the molecular condensate state. A more rigorous analysis has been performed in Ref. [23], using second-order correlation functions. However, it was limited to the case of a time-independent zero detuning between the atomic and molecular states, and deactivating collisions were not taken into account. As in the present analysis, Ref. [22] dealt with a time-dependent crossing of these states, in accordance with the experiments [19], and took into account deactivating collisions. We generalize here the parametric approximation, used in Refs. [12, 14] for the description of molecular dissociation into a single atomic mode, and in Ref. [11] for a multimode analysis of fluctuations in an unstable BEC.

The most impressive outcomes of the present approach are the extent of (near-total) conversion to entangled atoms or a molecular condensate, as well as the extreme degree of squeezing and the relatively long molecular BEC lifetimes achievable.

The model.— Consider a system of coupled atomic and molecular fields (see Ref. [20]) described by annihilation operators in the momentum representation \( \hat{\Psi}_a(\mathbf{p}, t) \) and \( \hat{\Psi}_m(\mathbf{p}, t) \), respectively. The coupling of the atomic and molecular fields (see Refs. [16, 17]) contains a product of two atomic creation operators and therefore de-
scribes the formation of entangled atomic pairs, in analogy with parametric down-conversion in quantum optics (see Ref. [3]). Spatial inhomogeneity due to the trapping potential and elastic collisions can be neglected here (see discussion below). Unlike the mean field used in Ref. [20], the atomic field is treated here as second-quantized, as in Ref. [13].

Let the initial state of the atomic field at \( t = t_0 \) be a coherent state of zero kinetic energy

\[
\hat{\Psi}_a (p, t_0) |\text{in}\rangle = (2\pi)^{3/2} \varphi_0 \delta(p) |\text{in}\rangle,
\]

where \( |\varphi_0|^2 = n_a(t_0) \) is the initial atomic density and \( |\text{in}\rangle \) is the time-independent state vector in the Heisenberg representation. A pair of condensate atoms forms a molecule of zero kinetic energy. Therefore the resonant molecules can be represented by a mean field \( \varphi_m(t) \), such that

\[
|\text{in}\rangle |\hat{\Psi}_m(p, t) |\text{in}\rangle = (2\pi)^{3/2} \varphi_m(t) \delta(p),
\]

where \( |\varphi_m(t)|^2 = n_m(t) \) is the molecular condensate density. This approach (unlike Refs. [2, 13]) takes into account the time dependence of the molecular mean field. Fluctuations of the molecular field due to collisions involving non-condensate atoms are neglected.

The outcome of atom-molecule and molecule-molecule deactivating collisions is introduced, as in Ref. [20], by adding molecular “dump” states. The elimination of these states in a second-quantized description is, however, different. It is similar to the Heisenberg-Langevin formalism in quantum optics (see Ref. [2]), but takes into account the nonlinearity of the collisional dumping. In the Markovian approximation, the equation of motion for the atomic field attains the form

\[
i\dot{\Psi}_a(p, t) = H \hat{\Psi}_a(p, t) + 2g^* \varphi_m(t) \hat{\Psi}_a(-p, t) + i \hat{F}(p, t)
\]

(3)

(using units with \( \hbar = 1 \), where

\[
H = \frac{p^2}{2m} - \mu B(t) - B_0 \frac{2}{2} - i\gamma |\varphi_m(t)|^2
\]

and \( m \) is the atomic mass. The second term in \( H \) describes the time-dependent Zeeman shift of the atom in an external magnetic field \( B(t) \), relative to half the energy of the molecular state, which is chosen as the zero energy, \( \mu \) is the difference in magnetic moments of an atomic pair and a molecule, and \( B_0 \) is the resonance value of \( B \). The coupling of the atomic and the molecular fields \( g \) is related to the phenomenological resonance strength \( \Delta \) as \( |g|^2 = 2\pi |a_\omega| \Delta / m \) (see Ref. [20]), where \( a_\omega \) is the elastic scattering length. The parameter \( \gamma \) describes the width of atomic states due to deactivating collisions (see Ref. [20]). The quantum noise source \( \hat{F}(p, t) \) provides conserving the correct commutation relations of the field operators.

Generalizing the parametric approximation [13], let us represent the atomic field operator in the form

\[
\hat{\Psi}_a(p, t) = \left[ \hat{A}(p, t) \psi_c(p, t) + \hat{A}^\dagger(-p, t) \psi_s(p, t) \right] \times \exp \left( -\int_{t_0}^t dt_1 \gamma |\varphi_m(t_1)|^2 \right).
\]

(5)

The operators \( \hat{A}(p, t) \) are expressible in terms of \( \hat{\Psi}_a(p, t_0), \hat{F}(p, t) \), and the \( c \)-number solutions \( \psi_{c,s}(p, t) \) of the equations

\[
i\dot{\psi}_{c,s}(p, t) = H \psi_{c,s}(p, t) + 2g^* \varphi_m(t) \psi_{c,s}^*(p, t),
\]

(6)

given the initial conditions \( \psi_c(p, t_0) = 1, \psi_s(p, t_0) = 0 \).

The atomic density

\[
n_a(t) = (2\pi)^{-3} \int d^3p_1 d^3p_2 \exp \left[ i (p_2 - p_1) \cdot r \right] \times |\langle \text{in}| \hat{\Psi}_a^\dagger(p_1, t) \hat{\Psi}_a(p_2, t) |\text{in}\rangle|, \]

(7)

then appears to be \( r \)-independent, and comprises the sum \( n_a(t) = n_0(t) + n_s(t) \) of the densities of condensate atoms \( n_0(t) = |\langle \text{in}| \hat{\Psi}_a(0, t) |\text{in}\rangle|^2 \), and of non-condensate (entangled) atoms \( n_s(t) \) in a wide spectrum of energies,

\[
n_s(t) = \int dE \tilde{n}_s(E, t), \]

(8)

where \( E \) is the kinetic energy of the non-condensate atoms.

The equation of motion for the molecular mean field \( \varphi_m(t) \) is obtained by a similar elimination of the dump fields from the corresponding operator equation, followed by a mean-field averaging. We thus obtain

\[
i\dot{\varphi}_m(t) = g m_a(t) - i \left( \gamma n_a(t) + \gamma_m |\varphi_m(t)|^2 \right) \varphi_m(t),
\]

(9)

where the parameter \( \gamma_m \) describes molecule-molecule deactivation collisions (see Ref. [20]). Here \( m_a \) is an anomalous density containing contributions of condensate and non-condensate atoms. The densities \( m_a, n_0, \) and \( \tilde{n}_s \) are all expressible in terms of \( \psi_c(p, t) \) and \( \psi_s(p, t) \). A numerical solution of Eqs. (3) on a grid of values of \( p \), combined with Eq. (8), is consistently sufficient for elucidating the dynamics of the system.

The present approach becomes mathematically equivalent to the approach of Ref. [22] if the inelastic collisions are neglected and the detuning is time-independent. At a low molecular density, the effect of non-condensate atoms is equivalent to the contribution to the width of the molecular state made in Ref. [20] for the same process.

Formation of a molecular condensate.— Calculations were performed for two Feshbach resonances in collisions of Na atoms, using parameter values presented in Ref. [24]. The strong resonance, at 907 G, has the
strength $\Delta = 0.98$ G, and the weak one, at 853 G, has the strength $\Delta = 9.5$ mG. The difference of magnetic momenta $\mu = 3.65$ (in Bohr magnetons), the elastic scattering length $a_e = 3.4$ nm, and the deactivation parameters are $\gamma = 0.8 \times 10^{-10}$ cm$^3$/s and $\tau_m = 10^{-9}$ cm$^3$/s. The neglection of elastic collisions is valid whenever $n_0 (t_0) \ll 10^{15}$ cm$^{-3}$ for the weak resonance, and $n_0 (t_0) \ll 10^{17}$ cm$^{-3}$ for the strong one. The spatial inhomogeneity can be neglected if the size of the condensate substantially exceeds $8 \times 10^{-2}$ cm$^{-1/2} \times n_0^{-1/2} (t_0)$ and $2.5 \times 10^{-2}$ cm$^{-1/2} \times n_0^{-1/2} (t_0)$, respectively, for the two resonances. Even when $n_0 (t_0) = 10^8$ cm$^{-3}$, these estimates set a minimal size of 8$\mu$m for the weak resonance and 2.5$\mu$m for the strong one. The variation of the magnetic field is linear in time, $B(t) = B_0 + Bt$.

A relatively long-lived molecular condensate is formed more effectively in the case of a backward sweep, when the molecular state crosses the atomic one downwards (see Fig. 1), as proposed in Ref. [21]. The maximal conversion efficiency of the atomic condensate to a molecular one is $2 \max (n_m) / n_0 \approx 0.8$ for the weak resonance (see Fig. 2). On increase of the atomic density, or on decrease of the ramp speed, the conversion efficiency falls due to inelastic collisions. On increase of the ramp speed, the probability of crossing to the molecular condensate decreases, leaving more atoms in the atomic condensate (see Ref. [20]). At low atomic densities the conversion becomes less efficient due to a temporary gain of population in the non-condensate atomic states. This observation is peculiar to, and emphasizes the importance of, the simultaneous consideration of inelastic collisions and molecular dissociation within the second-quantized approach. (The approach used in [20] does not describe the dissociation in the backward sweep.)

Figure 3 shows that a substantial conversion efficiency is retained in a wide region of the condensate density, leaving much freedom in the choice of the ramp speed appropriate for experiments. The lesser the initial density, the longer the lifetime $\tau_m$ of the molecular condensate but the higher the precision required for the control of the magnetic field.

The optimal ramp speed is approximately proportional to the initial density. This dependence minimizes the effect of a variation of parameters determining the conversion of the atomic condensate to the molecular one and loss of the molecular condensate. Indeed, the conversion to the molecular condensate is (in the fast decay approximation [20]) characterized by the parameter $g^2 n_0 / B$. Similarly, the loss is characterized by the ratio of the deactivation lifetime (which is inversely proportional to the initial density), and the crossing time (which is inversely proportional to the ramp speed).

The use of the strong resonance achieves a lower conversion efficiency, due to a gain in the temporary formation of non-condensate atoms. The optimal ramp speed is more than two orders of magnitude larger than in the weak resonance, given the same initial density.

**Formation of an entangled gas.—** As demonstrated in Ref. [13], the non-condensate atoms are formed in squeezed states, which now turn out to be two-mode squeezed states, as in Ref. [4]. It is similar to the state
The weak resonance in Na with the initial atomic density and the total atomic density (dotted line) calculated for FIG. 4: (a) Time dependence of the densities of the atomic condensate (dot-dashed line), entangled atoms (solid line), and the total atomic density (dotted line) calculated for the weak resonance in Na with the initial atomic density \( n_0 = 10^{14} \text{ cm}^{-3} \) and ramp speed 50 G/s in a forward sweep. The dashed line shows the mean squeezing parameter \( \bar{r} \) [see Eq. (10)]. (b) Energy spectra of the entangled-atom density \( \tilde{n}_s(E,t) \) (solid line) and the squeezing parameter \( r(E,t) \) (dashed line) calculated at the peak, \( t \approx -0.19 \) ms. The dotted and dashed lines show their values on the plateau at \( t \approx 4 \) ms.

of electromagnetic radiation formed by parametric down conversion. As in quantum optics Ref. [3], the amount of squeezing can be measured by the energy-dependent parameter \( r(E,t) \) related to the maximal and minimal uncertainties of quadratures involving creation and annihilation operators for opposite momenta [4]. A mean squeezing parameter, weighed by the spectral density of Eq. (8),

\[
\bar{r}(t) = \int dE \tilde{n}_s(E,t) r(E,t) / n_s(t).
\]  

is used to describe the time variation of the squeezing.

A stable gas of entangled atoms is formed by a forward sweep, in which the molecular state crosses the atomic one upwards (see Fig. 1). This process, too, is more efficient in the weak resonance. The molecular density is then very low and persists a shorter time (compared to that in the backward sweep) due to fast dissociation. Figure 4 demonstrates that more than 70% of the atomic condensate can be transformed into a gas of atoms in two-mode squeezed states with the mean squeezing parameter \( \bar{r} \approx 2.6 \), corresponding to a noise reduction of about 23 dB. The time dependence of the mean squeezing has a peak of \( \bar{r} \approx 3.1 \) at \( t \approx -0.19 \) ms. The state of an entangled gas can be frozen at the peak time by fast turning off of the magnetic field. The energy spectra of the entangled-atom density and the squeezing parameter are presented in Fig. 4b. The density spectra are rather narrow, and the peak energy increases with time. The squeezing parameter reaches the value of \( r(E,t) \approx 3.5 \) (noise reduction of 30 dB) at the energy \( E \approx 6 \text{ nK} \) and the time \( t \approx 4 \) ms.

Conclusions.— Both quantum corrections and deactivating collisions are necessary for the analysis of association in atomic BEC due to Feshbach resonance in a time-dependent magnetic field. Over 80% of the atomic population can be converted to a molecular condensate in a backward sweep. The molecules dissociate onto atoms in two-mode squeezed states that are entangled. In a forward sweep, very high squeezing may be obtained with the parameter \( r \) reaching a value of more than 3.

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