Coherent Molecular Solitons in Bose-Einstein Condensates

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We analyze the coherent formation of molecular Bose-Einstein condensate (BEC) from an atomic BEC, using a parametric field theory approach. We point out the transition between a quantum soliton regime, where atoms couple in a local way to a classical soliton domain, where a stable coupled-condensate soliton can form in three dimensions. This gives the possibility of an intense, stable atom-laser output. [S0031-9007(98)07283-4]

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Parametric solitons or simultaneous solitary waves ("simultons"), involving the optical $\chi^{(2)}$ nonlinearity, have been the topic of much recent theoretical and experimental interest in nonlinear optics. We propose a novel mechanism by which a similar phenomenon may occur in nonlinear atomic optics, in which coherent molecule formation in a Bose-Einstein condensate (BEC) takes the place of second harmonic generation.

This requires a coupling that converts two atoms into one molecule, thus generating coupled atomic and molecular Bose-Einstein condensates—and so taking advantage of molecular states that are known to exist in alkali-metal vapors. Our model includes a coherent molecular formation process (i.e., without dissipation) in an atomic BEC vapor [1] (or atom laser [2]), either through a Feshbach resonance [3] or Raman photoassociation [4]. We note that Feshbach resonances have already been observed [5]. The coherently coupled atom-molecular condensate could provide a route to the observation of a localized three-dimensional BEC soliton, even in the absence of a trap potential. A possible application is in the free propagation of a nondiverging atom-laser pulse, thus greatly increasing the intensity in an atom-laser beam. Even more than this would be the importance of observing the striking physical properties of this novel quantum field theory, and the corresponding Bose-enhanced chemical kinetics.

The original solution for the parametric soliton was in a one-dimensional environment [6]. These classical solutions have been classified topologically [7], and are generic to the mean-field theories of parametric nonlinearities that convert one particle into two (and vice versa). The equations are nonintegrable, and are different to the usual integrable classes of soliton equations. A considerable advantage of these types of nonlinear equations is that they are capable of providing solutions in one, two, or three space dimensions, which does not occur in the usual Gross-Pitaevskii equations. Both classical [6–8] and quantum [9] solutions have been recently identified (including observation of classical solitons in experiment [10]), although these different types of soliton have strikingly different qualitative behavior.

The purpose of this Letter is to point out the physical origin of these differences between the quantum and classical versions of the parametric field theory and to identify experimental requirements for observing these novel effects in Bose condensates. We consider the following basic Hamiltonian, to give a simple model of molecule formation:

$$\hat{H} = \hat{H}_0 + \hat{H}_1 + \hat{H}_{\rm int}\,, \tag{1}$$

where the free and interacting Hamiltonians are

$$\begin{aligned}
\hat{H}_0 &= \hbar \int d^3\mathbf{x} \left[\frac{\hbar}{2m} |\nabla\hat{\Phi}|^2 + \frac{\hbar}{2M} |\nabla\hat{\Psi}|^2 \right], \\
\hat{H}_1 &= \hbar \int d^3\mathbf{x} \left[\frac{\kappa}{2} \hat{\Phi}^{\dagger 2}\hat{\Phi}^2 + V_\Psi(\mathbf{x})\hat{\Psi}^\dagger\hat{\Psi} + V_\Phi(\mathbf{x})\hat{\Phi}^\dagger\hat{\Phi} \right], \\
\hat{H}_{\rm int} &= \hbar \int d^3\mathbf{x} \, \frac{\chi}{2} [\hat{\Phi}^2\hat{\Psi}^\dagger + \hat{\Phi}^{\dagger 2}\hat{\Psi}].
\end{aligned} \tag{2}$$

Here we define complex fields $\hat{\Phi} = \int d^3\mathbf{k}\hat{a}(\mathbf{k}) \times \exp[i(\mathbf{k}\cdot\mathbf{x})]$ and $\hat{\Psi} = \int d^3\mathbf{k}\hat{b}(\mathbf{k})\exp[i(\mathbf{k}\cdot\mathbf{x})]$. The field $\hat{\Phi}$ represents an atomic species of mass m in a potential $V_\Phi(\mathbf{x})$, in one internal state, while $\hat{\Psi}$ represents a dimer species of mass $M = 2m$, in a single vibrational and rotational state, with a potential $V_\Psi(\mathbf{x})$.

The coupling constant χ represents a formation rate for the dimer, in the S-wave scattering limit, while κ

represents the effective self-interaction of the atomic field. In the absence of any trap, the potentials are uniform, and $\hbar\rho = \hbar(V_\Psi - 2V_\Phi)$ is the formation energy of the dimer species. We note that these interactions are idealized, in the sense that both χ and κ represent processes that are microscopically nonlocal. To represent such nonlocal behavior, we must introduce a momentum cutoff k_m in the relative momenta of interacting fields, which physically must be around the inverse S-wave scattering length—if we wish to use the nonrenormalized effective potential to describe S-wave scattering. This is known to be essential to the correct interpretation of these types of effective field theories. It should be recognized that molecular self-interactions—as well as atom-molecular scattering—will occur as well. These are neglected here, since the relevant cross sections are not well known.

In the corresponding nonlinear optical case, the Φ and Ψ fields would correspond to a first and second harmonic, coupled by a $\chi^{(2)}$ nonlinearity of the dielectric, while κ would correspond to a $\chi^{(3)}$ nonlinearity. The interplay between quadratic and cubic nonlinearities in the case of nonlinear optical solitons has been analyzed, at the classical level and for one space dimension, in [11]. The effective masses, which should be different in the longitudinal and transverse directions, describe the effects of dispersion and diffraction, respectively, for both the fields (see, e.g., [9] for more details). Here the equations refer to a moving frame situation, with coordinates moving at the group velocity.

By comparison, in the directly comparable atomic case, we are considering atoms in free space. No potential needs to be included, since this is not essential to soliton formation. The molecular formation process would be tuned in any practical experiment, by magnetic fields or external Raman coupling, in order to reduce the energy mismatch $\hbar\rho$. An important consideration is the possible effects of losses due to inelastic atom-molecule collisions. We assume that an appropriate choice of molecular levels is made, so that these losses can be ignored over the relevant time scales for solitons to form. Thus, the neglect of molecular vibrational transitions is crucial to the present theory, which only includes one molecular level. An ideal situation would involve a direct coupling via a tuned Raman transition to the molecular ground state. A more sophisticated theory would include detailed atomic positions and multiple energy levels within each molecule. Our theory neglects these additional complications.

The Heisenberg equations of motion that correspond to the basic Hamiltonian are

$$i\frac{\partial}{\partial t}\hat{\Phi} = -\frac{\hbar}{2m}\nabla^2\hat{\Phi} + \chi\hat{\Psi}\hat{\Phi}^\dagger + \kappa\hat{\Phi}^\dagger\hat{\Phi}^2 + V_\Phi(\mathbf{x})\hat{\Phi}\,,$$
$$i\frac{\partial}{\partial t}\hat{\Psi} = -\frac{\hbar}{2M}\nabla^2\hat{\Psi} + \frac{\chi}{2}\hat{\Phi}^2 + V_\Psi(\mathbf{x})\hat{\Psi}\,. \qquad (3)$$

As a first step, we can take mean values, so that $\phi = \langle\hat{\Phi}\rangle$ and $\psi = \langle\hat{\Psi}\rangle$, and assume operator product factorization. This gives rise to mean-field equations, valid for a momentum cutoff less than the S-wave scattering length. For the case of Bose condensates in existing evaporative cooling experiments, near the atomic collective ground state, the mean-field equations represent modified Gross-Pitaevskii equations—which are known to successfully describe BEC excitations.

Another way to understand the behavior of this quantum many-body system is to look for energy eigenstates of the original Hamiltonian, in the limit of a large momentum cutoff. These must simultaneously be the eigenstates of $\hat{N} = \int d^3\mathbf{x}[|\hat{\Phi}|^2 + 2|\hat{\Psi}|^2]$, conserving the generalized particle number N (total number of atoms if we count each molecule as two atoms). Solving this, a remarkable fact emerges. We can show rigorously that in the limit of free space propagation, an N-boson ground state exists—by finding exact upper and lower bounds on the Hamiltonian energy. Since these coincide in three dimensions, we have the result that the (idealized) quantum ground-state energy is *exactly*

$$E_g^N = \frac{N}{2}\left(\hbar\rho - \frac{\hbar\chi^2}{2\kappa}\right), \qquad (4)$$

where we assume N is even. The proof of the lower bound also assumes $\kappa > 0$ and $\chi^2 > 2\rho\kappa$, and the result is obtained using the known solution of the two-particle ($N = 2$) bound-state problem [9].

This corresponds to $N/2$ independent quantum solitons or "dressed" molecules, each of which exist in a linear superposition with a pair of atoms (like a Cooper pair), so that

$$|\psi_Q^N\rangle = \left[\hat{b}^\dagger(0) + \int_0^{k_m} d^3\mathbf{k}\, g(\mathbf{k})\hat{a}^\dagger(\mathbf{k})\hat{a}^\dagger(-\mathbf{k})\right]^{N/2}|0\rangle\,. \qquad (5)$$

In this limit of a large cutoff in the quantum field theory, the ground-state energy has no lower bound as $\kappa \to 0$. This is in remarkable contrast to the known mean-field behavior of the corresponding classical energy, which is rigorously bounded below (see, e.g., [8]). Of more interest is the limiting behavior of the ground-state quantum energy when there is a cutoff k_m present. We have obtained a variational estimate of this quantity, and for this case we obtain ($\rho, \kappa \to 0$)

$$\tilde{E}_Q^N = -Nm\chi^2 k_m/(8\pi^2)\,. \qquad (6)$$

Here we have taken the case of a relatively large cutoff, so that the result assumes that $k_m \gg [\chi m/(2\pi\hbar)]^2$, and uses a variational ansatz of the form given previously. The ansatz gives us the true ground-state energy in the limit $k_m \to \infty$ (for any finite κ), since upper and lower energy bounds coincide. However, it is not necessarily the lowest possible energy at finite k_m. In order to show

this, we consider a coherent or mean-field ansatz, with broken symmetry, of form

$$|\psi_C^N\rangle = \exp\left\{\int d^3\mathbf{x}[\phi(\mathbf{x})\hat{\Phi}^\dagger(\mathbf{x}) + \psi(\mathbf{x})\hat{\Psi}^\dagger(\mathbf{x})]\right\}|0\rangle . \tag{7}$$

For this case, the classical decorrelation originates in coherent-state factorization properties of the Hamiltonian. This state is, however, not an eigenstate of $\hat{H}$ (since it is not an eigenstate of $\hat{N}$). It is an approximate (semiclassical) eigenstate at large N, and corresponds to two coupled Bose-Einstein condensates under broken symmetry conditions.

We will now show that, provided $\psi(\mathbf{x})$, $\phi(\mathbf{x})$ are chosen to minimize the classical Hamiltonian, they can give a lower energy than previously—although still finite. This calculation makes use of the known result that the classical parametric Hamiltonian is always bounded below [8], and the bound is given by the soliton energy for exact phase matching $\rho = 0$. This soliton energy is estimated by means of a variational ansatz applied to the Hamiltonian. We choose

$$\begin{aligned}\phi(\mathbf{x}) &= g_1 N^2[2/(\pi s_1)]^{3/4}\exp(-|\mathbf{x}|^2N^2/s_1),\\ \psi(\mathbf{x}) &= -g_2 N^2[2/(\pi s_2)]^{3/4}\exp(-|\mathbf{x}|^2N^2/s_2).\end{aligned} \tag{8}$$

The negative sign for $\psi(\mathbf{x})$ ensures that the coupling energy is negative, and the normalization implies that $g_1^2 + g_2^2 = 1$. We note that although a uniform variational ansatz is possible, it is known that a uniform field of this type is always unstable for a purely parametric coupling [12]—and hence cannot give the lowest energy.

Substituting into the Hamiltonian gives us the result

$$\begin{aligned}E_C^N/\hbar = N^3\left(\frac{3\hbar}{2m}\right)\left[\frac{g_1^2}{s_1} + \frac{g_2^2}{2s_2} - \frac{\tilde{\chi}g_1^2g_2s_2^{3/4}}{(s_1 + 2s_2)^{3/2}}\right]\\ + N^5\tilde{\kappa}g_1^4s_1^{-3/2} + N\rho g_2^2,\end{aligned} \tag{9}$$

where we have used $M = 2m$, with the simplified notation of $\tilde{\chi} = 2^{5/2}(2/\pi)^{3/4}m\chi/3\hbar$ and, similarly, $\tilde{\kappa} = 2^{-5/2}(2/\pi)^{3/2}\kappa$.

To minimize $\tilde{E}_C^N$, under the constraint of a fixed N, is a nontrivial algebraic procedure. However, the physics is considerably simplified in the region where the term in N^3 is dominant—which we note should not involve too large a contribution from the repulsive term that scales with N^5, and tends to destabilize soliton formation. In this region (i.e., assuming $\kappa \simeq \rho \simeq 0$), we obtain a coupled molecular Bose condensate minimum energy of

$$\tilde{E}_C^N = -CN^3\left(\frac{\hbar^2}{m}\right)\left(\frac{m\chi}{\hbar}\right)^4, \tag{10}$$

where C is a constant given by $C \simeq 1\cdot 2 \times 10^{-5}$. The relevant length scale is nearly identical for the two coupled condensates, and is given by

$$l_1 = \frac{\sqrt{s_1}}{N} \simeq 1.7\times 10^2\,\frac{1}{N}\left(\frac{\hbar}{m\chi}\right)^2. \tag{11}$$

This enables us to more clearly understand the apparent paradox that a full quantum theory gives a qualitatively different lower energy bound to the corresponding classical mean-field theory. To obtain a stable coupled atom-molecular condensate, we require $\tilde{E}_C^N \le \tilde{E}_Q^N$, which occurs at a critical boson number:

$$N \ge N_{\rm cr} = \sqrt{\frac{k_m}{8\pi^2 C}}\,\frac{\hbar}{m\chi}. \tag{12}$$

This question is therefore a subtle combination of momentum cutoff and particle density effects. To give some numerical results we consider $m \sim 10^{-25}$ kg, and use a χ-value estimate of about $\chi \sim 10^{-6}$ m$^{3/2}$/sec (given in [3], by Tommasini *et al.*, for a Feshbach resonance [5]), leading to $m\chi/\hbar \simeq 10^3$ m$^{-1/2}$. With a choice of the cutoff at $k_m \sim 1$ nm^{-1}, this gives a critical atom number of about $N_{\rm cr} \sim 10^3$, which is well within the range of current BEC experiments. At low particle density, the formation of individual dressed molecules is favored, as atoms couple to molecules in a particlelike way. The process is analogous to Rabi oscillations of atoms between two different electron sublevels, except that it occurs between pairs of atoms and the corresponding molecular levels. These dressed states have interesting properties, reminiscent of Cooper pairs, but cannot be described by the classical parametric soliton equations. At large couplings χ, and at large density (but not too large so that S-wave scattering is dominant) the coherent coupling of two entire condensates is dominant—just as in nonlinear optics. In this domain, provided other recombination processes are negligible, there are strong, coherent, and nonlinear wavelike interactions between the atomic and the molecular Bose condensates. For these parameters, it even appears possible to form a stable, three-dimensional, Bose-Einstein soliton.

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