PRL **94,** 190402 (2005) PHYSICAL REVIEW LETTERS week ending 20 MAY 2005

Complex Chemical Potential: Signature of Decay in a Bose-Einstein Condensate

George E. Cragg[1] and Arthur K. Kerman[2]

[1]*Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*
[2]*Center for Theoretical Physics, Laboratory for Nuclear Science and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*

(Received 5 May 2004; published 19 May 2005)

We explore the zero-temperature statics of an atomic Bose-Einstein condensate in which a Feshbach resonance creates a coupling to a second condensate component of quasibound molecules. Using a variational procedure to find the equation of state, the appearance of this binding is manifest in a collapsing ground state, where only the molecular condensate is present up to some critical density. Further, an excited state is seen to reproduce the usual low-density atomic condensate behavior in this system, but the molecular component is found to produce a coherent, many-body decay, quantified by the imaginary part of the chemical potential. Most importantly, the unique decay rate dependencies on density ($\sim\rho^{3/2}$) and on scattering length ($\sim a^{5/2}$) can be measured in experimental tests of this result.

DOI: 10.1103/PhysRevLett.94.190402 PACS numbers: 03.75.Hh, 02.30.Xx, 05.30.Jp, 32.80.Pj

Since the production of atomic Bose-Einstein condensates (BEC) in the laboratory [1,2], many schemes have been proposed whereby the experimentalist may control the interatomic interactions governing the behavior of these gases [3]. One such proposal involves a Feshbach resonance in which two atoms combine to form a quasibound molecule [4,5]. This molecule is described as the intermediate state or closed channel of the scattering reaction as the constituent atoms in general have different spin configurations in the bound state than in the scattering state. Because of its dependence on the internal spin states, the energy difference, or detuning between the scattered and bound states can thus be tuned using the Zeeman effect in an external magnetic field. As the binding energy of the molecular state is brought close to the energy of the colliding atoms, the appearance of these loosely bound molecules increases. Consequently, the coupling between atoms and molecules acts to modify the effective interatomic interactions. Near zero energy, these interactions are described by the s-wave scattering length, which in turn can be tuned by varying the external magnetic field. This degree of control suggests that initially unstable condensates may be rendered stable by adjusting the scattering length from negative to positive values. We provide a many-body variational description of such a scenario, specifically using the case of ^{85}Rb to compare our results with experiment [6,7]. For a uniform system our findings reveal a collapsing ground state and a decaying excited state, where the latter fits the behavior seen from experiment. However, in the excited state, a complex chemical potential is obtained where the imaginary part determines the decay rate.

Before embarking on the many-body analysis of the coupled atom-molecule BEC, it is first necessary to review the relevant two-body physics underlying the interatomic interactions. In particular, the two-body formalism provides a means of replacing the interaction strength by the s-wave scattering length as the relevant parameter in the many-body equations. In two-atom scattering, there are a number of different channels or outcomes of the scattering event, corresponding to both the closed and open channels of the bound and scattering states, respectively. Because coupling will be strongest to the highest lying molecular state below the continuum of scattering states, we consider a two-channel model with coupling between a single open and a single closed channel. Described in detail elsewhere [4], the coupled channels approach is only summarized here. Taking a separable form for the two-body pseudopotential we have,

$$\langle \mathbf{k}|V|\mathbf{k}'\rangle = \lambda v(\mathbf{k})v(\mathbf{k}'), \tag{1}$$

where $\mathbf{k}$ is the momentum in the center of mass and λ is the interaction strength, which is taken to be negative, since we assume an attractive background throughout. Using the separable potential with a molecular form factor equal to $v(\mathbf{k})$, the coupled channels analysis yields an integral equation for the two-body wave function $\Psi(\mathbf{k})$ [we use the notation $\int_{\mathbf{k}} = (2\pi)^{-3}\int d^3k$],

$$(2\mathbf{k}^2 - E)\Psi(\mathbf{k}) + \left(\lambda - \frac{\lambda^2\alpha^2}{\epsilon - E}\right)v(\mathbf{k})\int_{\mathbf{k}'} v(\mathbf{k}')\Psi(\mathbf{k}') = 0, \tag{2}$$

where E, α, and ϵ are the center of mass energy, coupling strength parameter, and detuning, respectively. Here and throughout, all energies are on the scale of $\hbar^2/2m$. Equation (2) is recognized as the usual single channel two-body integral scattering equation [8], where the molecular coupling has replaced the background interaction strength λ with an energy dependent form, $\lambda - \lambda^2\alpha^2/(\epsilon - E)$.

Starting with the relationship between the scattering length a and the T matrix $8\pi a = \langle \mathbf{k}|T|\mathbf{k}'\rangle|_{\mathbf{k}=\mathbf{k}'=0}$, we derive an equation between the interaction strength and

© 2005 The American Physical Society

the range. To solve for the separable T matrix $[\langle \mathbf{k}|T|\mathbf{k}'\rangle = v(\mathbf{k}) t v(\mathbf{k}')]$, Eq. (1) is substituted into the Lippmann-Schwinger equation, $T = V + VGT$, where G is the free particle Green's function. Subsequently, the background scattering length is obtained as

$$\frac{1}{8\pi a_{\mathrm{bg}}} = \frac{1}{\lambda} + \frac{1}{b}, \tag{3}$$

where the range is defined by $b = 1/\int_{\mathbf{k}} v(\mathbf{k})^2/2k^2$ [9]. As found from Eq. (2), we replace the background strength λ by $\lambda - \lambda^2\alpha^2/(\epsilon - E)$ to include the effect of molecular coupling. With $E = 0$, this substitution gives the full scattering length

$$a(\epsilon) = \frac{\epsilon a_{\mathrm{bg}}}{\epsilon + 8\pi a_{\mathrm{bg}}\alpha^2}, \tag{4}$$

where we have taken the zero range limit in which $v(\mathbf{k}) \rightarrow 1 \Rightarrow b \rightarrow 0$ and $\lambda \rightarrow 0^-$ such that a_{bg} remains finite. Alternatively, the full scattering length can be expressed in terms of the external magnetic field B since the Zeeman detuning is given as $\epsilon = \gamma(B - B1)$, with γ being the atomic species dependent proportionality and with $B1$ as the offset field. These two-body results will be used to simplify the many-body treatment.

We begin with a many-body Hamiltonian that describes an infinite, uniform medium, as subsequent results can be applied to finite systems by way of the local density approximation. Hence, the model Hamiltonian includes the atomic kinetic energy, atom-atom collisions, the molecular detuning energy, and a coupling between atoms and molecules. Replacing the usual two-body term $\hat{\psi}^4$ with an auxiliary field $\hat{\chi}$, coupled to the atomic field $\hat{\psi}$ through a single Feshbach resonance, obtains a simplified expression that is quadratic in the atomic field instead of quartic [10,11]:

$$\begin{aligned}
\hat{H} - \mu\hat{N} = {} & \sum_{\alpha\beta}\hat{\psi}^\dagger_\alpha(p^2_\beta - \mu)\delta_{\alpha\beta}\hat{\psi}_\beta + (\epsilon - 2\mu)\sum_\alpha \hat{\phi}^\dagger_\alpha\hat{\phi}_\alpha \\
& + (\varepsilon - 2\mu)\sum_\alpha \hat{\chi}^\dagger_\alpha\hat{\chi}_\alpha \\
& + \frac{\lambda\alpha}{\sqrt{2}}\sum_{\alpha\beta\gamma}(\hat{\phi}^\dagger_\alpha F_{\alpha\beta\gamma}\hat{\psi}_\gamma\hat{\psi}_\beta + \mathrm{H.c.}) \\
& + \frac{g}{\sqrt{2}}\sum_{\alpha\beta\gamma}(\hat{\chi}^\dagger_\alpha F_{\alpha\beta\gamma}\hat{\psi}_\gamma\hat{\psi}_\beta + \mathrm{H.c.}).
\end{aligned} \tag{5}$$

In anticipation of the variational procedure to be carried out, the grand canonical form is used with the usual identification of the Lagrange multiplier as the chemical potential. Furthermore, the atomic field is coupled to both the molecular field [12] $\hat{\phi}$ and the auxiliary field $\hat{\chi}$ with coupling constants $(\lambda\alpha, g)$ and detunings (ϵ, ε) implicitly defined in Eq. (2), where the background atom-atom interaction is reproduced by setting $g^2/(\varepsilon - E) = -\lambda$ with $|\varepsilon| \gg |E|$. In the zero range limit $(\lambda \rightarrow 0^-)$, these conditions are satisfied by setting $\varepsilon = 2/\lambda^2$ and $g = \sqrt{-2/\lambda}$. Without loss of generality, zero range also allows us to take the same molecular form factor F setting it equal to the v defined in the separable potential of Eq. (1).

For the zero-temperature analysis of the Hamiltonian, we work in the Schrödinger picture using a static, "squeezed" Gaussian trial wave functional [13] given by

$$\begin{aligned}
\Psi[\psi, \phi, \chi] = {} & N_\psi \exp\left\{-\sum_{\alpha\beta}\delta\psi'_\alpha\frac{1}{4}G^{-1}_{\alpha\beta}\delta\psi'_\beta\right\} \\
& \times N_\phi \exp\left\{-\sum_\alpha\frac{1}{2}\delta\phi'^2_\alpha\right\} N_\chi \exp\left\{-\sum_\alpha\frac{1}{2}\delta\chi'^2_\alpha\right\},
\end{aligned} \tag{6}$$

where N_ψ, N_ϕ, and N_χ are normalization constants, and the width G is taken as real. Denoting ψ' as the atomic field with ψ as its mean, the atomic field fluctuations are given as $\delta\psi' = \psi' - \psi$, with the analogs of the molecular fluctuations defined similarly. In the momentum space representation of the uniform system, all fields assume their mean values times a Dirac delta function $\delta(\mathbf{k})$, whereas the Gaussian width assumes the diagonal form $G(\mathbf{k})^{-1}\delta(\mathbf{k} - \mathbf{k}')$. Taking the expectation value of Eq. (5) with respect to the trial functional of Eq. (6) gives an expression for the pressure, $-P = \langle\hat{H} - \mu\hat{N}\rangle/V$, in a volume V. Stationarizing this quantity in $G(\mathbf{k})$, and in the mean fields ψ, ϕ, and χ, gives a set of variational equations, which in momentum space can be expressed as

$$G(\mathbf{k}) = \frac{1}{2}\sqrt{\frac{k^2 - \mu + \xi}{k^2 - \mu - \xi}}, \tag{7}$$

$$(\xi - \mu)\psi = 0, \tag{8}$$

$$-\frac{\xi}{8\pi a(\epsilon - 2\mu)} + \int_{\mathbf{k}}\left[D(\mathbf{k}) + \frac{\xi}{2k^2}\right] + \frac{1}{2}\psi^2 = 0, \tag{9}$$

where $\xi = \lambda\alpha\phi + g\chi$. Arriving at Eq. (9) requires the use of Eqs. (3) and (4), replacing ϵ by $\epsilon - 2\mu$. Additionally, we find that the atomic density $\langle\hat{\psi}^\dagger\hat{\psi}\rangle$ and the anomalous atomic density $\langle\hat{\psi}\hat{\psi}\rangle$ are given by $\int R(\mathbf{k}) + \psi^2/2$ and $\int D(\mathbf{k}) + \psi^2/2$, respectively, where the pair correlations $R(\mathbf{k})$ and $D(\mathbf{k})$ are related through the Gaussian width as

$$R(\mathbf{k}) = \frac{1}{2}\left[\frac{1}{4G(\mathbf{k})} + G(\mathbf{k}) - 1\right], \tag{10}$$

$$D(\mathbf{k}) = \frac{1}{2}\left[\frac{1}{4G(\mathbf{k})} - G(\mathbf{k})\right]. \tag{11}$$

From Eqs. (3), (4), (7), (10), and (11), an expression for the pressure is obtained,

$$-P=-\frac{\xi^2}{16\pi a(\epsilon-2\mu)}+\frac{1}{2}(\xi-\mu)\psi^2+\int_{\mathbf{k}}\frac{1}{2}\left[\sqrt{(k^2-\mu)^2-\xi^2}-(k^2-\mu)+\frac{\xi^2}{2k^2}\right], \tag{12}$$

which can then be differentiated to find the number density,

$$\rho=\frac{\partial P}{\partial\mu}=\int_{\mathbf{k}}R(\mathbf{k})+\frac{1}{2}\psi^2+\frac{\alpha^2\xi^2}{(\epsilon-2\mu)^2}, \tag{13}$$

thus showing the contributions from the correlated atom pairs as well as the two condensates.

We begin the analysis of these solutions by noticing that for a nonzero ξ in Eq. (7), G is real only when μ is taken negative, in contrast to the usual positive chemical potential $8\pi a\rho$ of stable BECs. From Eq. (8) we see that there are two solutions, with one given by a vanishing atomic field ($\psi = 0$), whereas the other is given by a nonvanishing field and $\xi = \mu < 0$. Considering the $\psi = 0$ solution first, Eq. (11) is substituted into Eq. (9), resulting in an expression that determines μ in terms of ξ. We find that this equation has a solution for ξ from zero only up to some critical value ξ_c. From Eq. (13), this interval corresponds to a range of densities, starting from zero with ξ and ending at some critical value ρ_c, corresponding to ξ_c. As shown in Fig. 1, the plot of the energy per particle $e = \int(P/\rho^2)d\rho$ for this solution results in a downward curve starting at half the molecular binding energy BE/2 and stopping at ρ_c. To find the remaining solution for $\rho > \rho_c$, we take $\xi = \mu$ in Eq. (8), and then use Eq. (9) to solve for $\psi^2/2$. In this case, the energy per particle starts at ρ_c, then continues downward for arbitrarily large ρ, (see Fig. 1). Thus, these two solutions comprise a single curve of two pieces, recognized as the collapsing ground state of the model Hamiltonian [14].

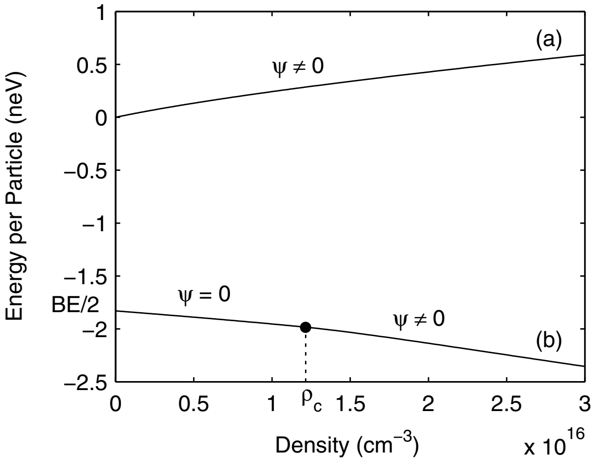

FIG. 1. The real part of the excited state (*a*) and the collapsing ground state (*b*) are evaluated using the following numerical values for ^{85}Rb: applied magnetic field, $B = 162.3$ G [6], offset field $B1 = 165.6$ G, coupling constant $-8\pi a_{\mathrm{bg}}^3\alpha^2 = 330$, detuning proportionality $\gamma a_{\mathrm{bg}}^2 = -30\ \mathrm{G}^{-1}$, and the respective background and full scattering lengths $a_{\mathrm{bg}} = -450a_0$ and $a(\epsilon) \approx 193a_0$, where a_0 is the Bohr radius [19]. With these values, we obtain a critical density, ρ_c, of $1.22 \times 10^{16}\ \mathrm{cm}^{-3}$ and a BE of -3.7 neV. As the applied magnetic field is decreased toward resonance, ρ_c decreases, thereby shortening the range of the $\psi = 0$ solution. Tuning the scattering length to negative values results in a collapsing atomic-molecular condensate solution, with the usual low-density dependence, $e \sim 4\pi a(\epsilon)\rho$.

Thus far, we have not uncovered the solution corresponding to the low-density behavior $e \sim 4\pi a(\epsilon)\rho$ reported in the experiments for the positive scattering length regime of ^{85}Rb [6]. Obtaining this solution requires a closer examination of the density expression for $\xi = \mu$. Along with the zero-range limit, we use Eqs. (7) and (9)–(11), to express the right-hand side of Eq. (13) in terms of the chemical potential μ;

$$\rho=\frac{\sqrt{2}}{3\pi^2}(-\mu)^{3/2}+\mu\left(\frac{\alpha^2}{\epsilon-2\mu}+\frac{1}{8\pi a_{\mathrm{bg}}}\right)+\frac{\alpha^2\mu^2}{(\epsilon-2\mu)^2}. \tag{14}$$

Since Eq. (14) has no solution for both μ, $\rho > 0$, we expand in $\sqrt{\rho}$, which is constrained to be real and positive,

$$\mu=8\pi a(\epsilon)\rho-i\frac{\sqrt{\pi}}{3}256a(\epsilon)^{5/2}\rho^{3/2}+\ldots. \tag{15}$$

Thus, this expansion is seen to give the desired low-density $4\pi a(\epsilon)\rho$ dependence of the energy per particle, but becomes complex at higher order [15].

At first glance, the complex-valued chemical potential appears unphysical. However, the chemical potential can also play the role of phase of the atomic field, as can most easily be seen from the Heisenberg equation for ψ, $i\hbar\partial\psi^*/\partial t = \langle[\hat{\psi}^\dagger, \hat{H}]\rangle = -\xi(t)^*\psi(t)$, in which the expectation value is carried out using the trial wave functional of Eq. (6). Consistency with the variational analysis requires taking the time dependence of the fields as $\psi(t) = \psi\exp(-i\mu t/\hbar)$ and $\xi(t) = \xi\exp(-2i\mu t/\hbar)$, which is then seen to yield the variational ψ equation (8).

From this vantage point, the imaginary part of μ is seen to be proportional to the decay rate. The inspiration for such an interpretation arises in the context of QED, where a complex action signifies decay of a constant, uniform electric field [16]. Using the ^{85}Rb parameters given in Fig. 1 and taking $\sim 10^4$ atoms in a BEC cloud of radius 25 μm [6], a decay constant τ of approximately 14 s is obtained to lowest order in $\rho[\tau \sim 1/\rho^{3/2}]$ [17]. The full solution of Eq. (14) leads to a seventh order equation in $\sqrt{-\mu}$, which, when solved numerically, gives a decay constant of approximately 14.2 s, thus confirming the validity of the expansion in Eq. (15). Hence, this result is in qualitative agreement with the 10 s decay reported in Ref. [6], where the experiments were carried out in a regime where this decay process dominates over the two and three-body inelastic processes [18]. As seen from

Eq. (15), this decay rate has novel density and scattering length dependencies $[\sim a(\epsilon)^{5/2}\rho^{3/2}]$, which can be experimentally tested.

Finally, we address the physical interpretations implied by this theory. Because of the fixed particle number, the decay of the excited state does not represent atom loss, but is instead ascribed to a transition into collective phonon excitations of the lower state. Using a small oscillation analysis, this statement is confirmed by the fact that the excited solution lies in a continuum of the collapsing state modes. Therefore, the coherent decay can be viewed as a precursor to the usual inelastic loss mechanisms present in the collapse to higher density. Additionally, it may be questioned what meaning can be assigned to other thermodynamic quantities derived from a complex chemical potential. In such cases, the real part of these quantities acquire the usual physical interpretation, where the imaginary pieces are seen as signatures of the decay.

In summary, we have used a variational procedure in extremizing a many-body Hamiltonian describing an atom-molecule BEC coupled through a Feshbach resonance. For positive scattering lengths, we have found that the existence of the bound molecular state results in a collapsing lower state and an excited, "false vacuum" with a complex-valued chemical potential. By examining the Heisenberg equation for the atomic field, the imaginary part of the chemical potential is seen to physically correspond to the decay rate. Since the excited state lies inside the continuum of phonon excitations of the collapsing lower state, the decay corresponds to a transition of the system into these collective excitations. Moreover, the unique scattering length and density dependencies would allow conclusive verification of this coherent, many-body process. Finally, the associated complex thermodynamic quantities have imaginary parts that are simply signatures of the decay, whereas the real parts assume their usual physical meaning.

We thank Eddy Timmermans for many useful discussions. This work was supported by the MIT-Los Alamos Collaborative Research Grant to Develop an Understanding of Bose-Einstein Condensates, Contract No. 19442-001-99-35.

---

[1] M. H. Anderson *et al.*, Science **269**, 198 (1995).
[2] K. B. Davis *et al.*, Phys. Rev. Lett. **75**, 3969 (1995).
[3] Many other methods involve applying external laser fields, see P. W. Milonni and A. Smith, Phys. Rev. A **53**, 3484 (1996); P. S. Julienne, K. Burnett, Y. B. Band, and W. C. Stwalley, *ibid.* **58**, R797 (1998); P. O. Fedichev, Yu. Kagan, G. V. Shlyapnikov, and J. T. M. Walraven, Phys. Rev. Lett. **77**, 2913 (1996).
[4] The theoretical description here is in analogy with that for nuclear reactions as described by H. Feshbach, Ann. Phys. (N.Y.) **281**, 519 (2000).
[5] For experimental work on Feshbach resonances see, for example, S. Inouye *et al.*, Nature (London) **392**, 151 (1998); T. Volz *et al.*, Phys. Rev. A **68**, 010702 (2003).
[6] S. L. Cornish *et al.*, Phys. Rev. Lett. **85**, 1795 (2000).
[7] J. L. Roberts *et al.*, Phys. Rev. Lett. **86**, 4211 (2001); E. A. Donley, N. R. Claussen, S. T. Thompson, and C. E. Wieman, Nature (London) **417**, 529 (2002).
[8] E. Merzbacher, *Quantum Mechanics* (Wiley, New York, 1998), p. 534.
[9] An equivalent renormalization can be found in S. J. J. M. F. Kokkelmans and M. J. Holland, Phys. Rev. Lett. **89**, 180401 (2002); S. J. J. M. F. Kokkelmans *et al.*, Phys. Rev. A **65**, 053617 (2002).
[10] Use of the auxiliary molecular field is analogous to the Hubbard-Stratonovich transformation as found in S. Weinberg, *The Quantum Theory of Fields I* (Cambridge University Press, Cambridge, England, 1995), p. 461.
[11] The Heisenberg equations derived from a quartic Hamiltonian are given in the Phys. Rev. Lett. of Ref. [9]; M. Holland, J. Park, and R. Walser, Phys. Rev. Lett. **86**, 1915 (2001). We remind the reader that such model Hamiltonians do not include the effects of deeply bound molecular states, usually addressed by discussions of three-body loss rates.
[12] Coupling to a molecular field was first put forth in P. Tommasini *et al.*, cond-mat/9804015; E. Timmermans *et al.*, cond-mat/9805323; E. Timmermans, P. Tommasini, R. Côté, M. Hussein, and A. Kerman, Phys. Rev. Lett. **83**, 2691 (1999).
[13] A complete description of the variational procedure used here is given by A. K. Kerman and P. Tommasini, Ann. Phys. (N.Y.) **260**, 250 (1997).
[14] An entire series of collapsing solutions is obtained below curve (b) if additional resonances to more tightly bound states are included in the Hamiltonian. Although each of these proceeds toward collapse, an appropriate finite range atomic potential must be used in the Hamiltonian for it to describe the high-density liquid.
[15] Although the hard-core Bose gas also has the first two terms depending only on the scattering length [see A. L. Fetter and J. D. Walecka, *Quantum Theory of Many-Particle Systems* (Dover, New York, 2003), Eq. (22.19), p. 222], the dependence differs in the second term since the hard-core potential admits no bound state.
[16] J. Schwinger, Phys. Rev. **82**, 664 (1951).
[17] To obtain a good approximation to τ we simply equated the density to $N/[(4/3)\pi r^3]$, but a consistent local density approximation includes $\mu\psi$ in the Gross-Pitaevskii equation, thereby producing a complex self-consistent potential.
[18] J. L. Roberts, N. R. Claussen, S. L. Cornish, and C. E. Wieman, Phys. Rev. Lett. **85**, 728 (2000).
[19] Comparing Eq. (4) with the form $a(B) = a_{\rm bg}[1 - \Delta/(B - B_0)]$, we find $\Delta = 8\pi a_{\rm bg}\alpha^2/\gamma$ and $B1 = B_0 + \Delta$, where the values for Δ, B_0, and $a_{\rm bg}$ can be found in the Nature (London) citation of Ref. [7]. The difference in magnetic moments between open and closed channels, quoted in Fig. 1 of the Phys. Rev. Lett. of Ref. [9], is then used to find γ.