Rarified Liquid Properties of Hybrid Atomic-Molecular Bose-Einstein Condensates

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In the atomic Bose-Einstein condensate, the interactions that bring a binary atom system to an intermediate state molecule in the Feshbach resonance create a second condensate component of molecules. The atomic and molecular condensates coherently exchange pairs of atoms. We discuss a signature of the coherent intercondensate exchange: Josephson-like oscillations of the atomic and molecular populations in response to a sudden change of the energy detuning. The dependence of the many-body ground state energy on volume suggests that the on-resonant ground state is a dilute condensate with the liquidlike property of a self-determined density.

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As dilute gases, the atomic Bose-Einstein condensates [1–3] are amenable to atomic manipulation techniques. As superfluids [4], these dilute condensates exhibit an unusual degree of flexibility, promising novel studies of macroscopic quantum coherence. For instance, the notion that external fields can alter the interatomic interactions [5] suggests the study of condensates with interparticle interactions that can be experimentally controlled [6]. Signatures of one of the proposed schemes, the low energy Feshbach resonance [7], have been observed recently [8]. In this Letter, we point out that this resonance affects the condensate system more profoundly than altering an effective interparticle interaction: the molecules, formed in the intermediate state of the binary atom resonance, occupy a second condensate component. The atomic and molecular condensates interact in part by coherently exchanging pairs of atoms, implying interesting and unusual properties. Below, we discuss two examples: (i) The molecular condensate, even if it is small in the off-resonant regime, can reveal its presence by means of Josephson-like oscillations of the atomic and molecular populations in response to a sudden change of the detuning. (ii) Close to resonance, the coherent intercondensate tunneling binds the dilute many-body Bose-Einstein condensate (BEC) to a system with the liquidlike property of a self-determined density.

In the low energy Feshbach resonance, the hyperfine interaction, V_{hf}, rearranges the spins of two alkali atoms interacting in an external magnetic field $\mathbf{B}$, bringing them to an intermediate quasibound molecular state. In this process, a valence electron spin is "flipped" which, in the magnetic field, raises the continuum of the intermediate spin state $|S'\rangle$ by an amount $\Delta(B)$, relative to the continuum of the initial binary atom spin state $|S\rangle$. At a resonant magnetic field $B = B_m$, the $|S\rangle$ continuum lines up with the bound state m of the $|S'\rangle$ interatomic potential. Near the resonance, the energy difference ϵ of the initial and intermediate states, which we call "detuning," varies with magnetic field, $\epsilon \approx (\partial\Delta/\partial B) \times [B - B_m]$. The amplitude of the resonant process is characterized by $\alpha = \langle S'|V_{hf}|S\rangle \times \int d^3r\, \varphi_m^*(\mathbf{r})\varphi(\mathbf{r})$, where φ_m denotes the vibrational wave function of the m state and φ the continuum ($|S\rangle$) wave function. In the relevant low energy limit, α is independent of energy and of B (to the extent that $|S\rangle$ and $|S'\rangle$ are B independent).

The resonance width is proportional to the phase space volume available to the binary atom final state. As this volume vanishes in the condensate limit of zero relative velocity, the width vanishes, and the binary atom effective scattering length (a without resonance) depends dispersively on the detuning: $a_{\text{eff}} = a - (m/4\pi\hbar^2)\alpha^2/\epsilon$. On resonance, $\lim_{\epsilon\to 0}$, the effective scattering length diverges, and simply replacing the scattering length by a_{eff} in the dilute single condensate expressions leads to unphysical divergencies of quantities such as pressure and chemical potential.

Rather than starting from the *a priori* assumption of an effective scattering length, we include the long-lived molecules explicitly in the many-body theory [9]. The atom-molecule coupling contributes the following term to the second-quantized Hamiltonian:

$$\hat{H}_{\text{at-mol}} = \frac{\alpha}{\sqrt{2}} \int d^3r\, \{\hat{\psi}_m^\dagger(\mathbf{r})\hat{\psi}_a(\mathbf{r})\hat{\psi}_a(\mathbf{r}) + \text{H.c.}\}, \quad (1)$$

where $\hat{\psi}_a$ and $\hat{\psi}_m$ denote the field operators of the atoms and the quasibound molecules, respectively. In describing binary atom collisions, Eq. (1) reproduces the correct resonant behavior. In the many-body system, we account for the elastic atom-atom, molecule-molecule, and atom-molecule interactions by means of the corresponding interaction strengths, λ_a, λ_m, and λ [10]. Measuring energy relative to that of noninteracting atoms, the detuning ϵ

makes its appearance as the single molecule energy, and the many-body Hamiltonian is

$$\begin{aligned}\hat H = {}& \int d^3r\, \hat\psi_a^\dagger \left[-\frac{\hbar^2\nabla^2}{2m} + \frac{\lambda_a}{2}\, \hat\psi_a^\dagger \hat\psi_a + \lambda \hat\psi_m^\dagger \hat\psi_m \right] \hat\psi_a \\ &+ \int d^3r\, \hat\psi_m^\dagger \left[-\frac{\hbar^2\nabla^2}{4m} + \frac{\lambda_m}{2}\, \hat\psi_m^\dagger \hat\psi_m + \epsilon \right] \hat\psi_m \\ &+ \frac{\alpha}{\sqrt{2}} \int d^3r\, \{ \hat\psi_m^\dagger \hat\psi_a \hat\psi_a + \hat\psi_m \hat\psi_a^\dagger \hat\psi_a^\dagger \}, \qquad (2)\end{aligned}$$

where the position dependence of the field operators is understood. Bose-Einstein condensation is generally characterized by a nonvanishing order parameter, $\langle \hat\psi \rangle = \phi \neq 0$, which we refer to as condensate field. In a mean-field approach, we obtain the equation of motion of the atomic ($\phi_a = \langle \hat\psi_a \rangle$) and molecular ($\phi_m = \langle \hat\psi_m \rangle$) fields by taking the expectation value of the Heisenberg equations, $i\hbar \dot{\hat\psi}_a = [\hat\psi_a, \hat H]$, $i\hbar \dot{\hat\psi}_m = [\hat\psi_m, \hat H]$. Under the assumption of complete coherence, e.g., $\langle \hat\psi_a \hat\psi_a \rangle \approx \phi_a^2$, appropriate for the dilute zero temperature condensates, the expectation value of the Heisenberg equations yields coupled nonlinear equations of motion that replace the single-condensate Gross-Pitaevskii equation [11–13]:

$$\begin{aligned} i\hbar \dot\phi_a &= \left[-\frac{\hbar^2\nabla^2}{2m} + \lambda_a n_a + \lambda_m n_m \right] \phi_a + \sqrt{2}\, \alpha\, \phi_m \phi_a^*, \\ i\hbar \dot\phi_m &= \left[-\frac{\hbar^2\nabla^2}{4m} + \epsilon + \lambda_m n_m + \lambda n_a \right] \phi_m + \frac{\alpha}{\sqrt{2}}\, \phi_a^2. \end{aligned} \qquad (3)$$

In Eq. (3), n_a and n_m denote the atomic and molecular condensate densities: $n_a = |\phi_a|^2$ and $n_m = |\phi_m|^2$. The α coupling terms describe coherent intercondensate exchange of atom pairs. Note that the condensate coherence of the fields is crucial in obtaining Eq. (3)—if not for the atomic condensate, $\langle \hat\psi_a \hat\psi_a \rangle = 0$.

Current experiments resonate on molecular states of high vibrational quantum number v. Consequently, interactions that change the molecular state (decreasing v) are particularly relevant. The "fragility" of these molecules is expressed by high rate constants $c_{m,a}$ and $c_{m,m}$ for state-changing collisions with atoms and other molecules (estimates [14] give $c_{m,a} \sim 10^{-9}$–10^{-11} cm^3 s^{-1}, as compared to $c_{a,a} \sim 10^{-14}$ cm^3 s^{-1} for binary atom state changing collisions). For noncondensed systems, the loss processes can be accounted for by a rate equation, $\dot n_m = -[c_{m,a} n_a + c_{m,m} n_m] n_m$. For BEC systems, the same collisions affect the coherent dynamics. A lowest-order perturbation treatment modifies the $\dot\phi_m$ equation by rendering the interaction strengths absorptive: $\lambda \rightarrow \lambda - i\hbar c_{m,a}/2$ and $\lambda_m \rightarrow \lambda_m - i\hbar c_{m,m}/2$.

In the off-resonant limit, which we define here as $\epsilon \gg \lambda n$ and $\epsilon \gg \lambda_m n$, where n denotes the atomic condensate particle density, $n = n_a + 2n_m$, the molecule fraction is small and the state-changing molecular collisions destroy the condensate slowly. In spite of the smallness of the molecular condensate, the dynamical response to a sudden change of the magnetic field strength (i.e., detuning) carries a detectable signature of the condensate: Josephson-like oscillations of the atomic and molecular populations. In this limit, propagation of the atomic condensate is hardly affected by the molecular condensate: $\phi_a \approx \sqrt{n} \exp(-i\lambda_a n t/\hbar)$. In this approximation, the molecular field of a homogeneous system evolves according to

$$\begin{aligned} i\hbar \dot\phi_m = {}& \left[\epsilon(t) + \lambda n - i\, \frac{\gamma_m}{2} \right] \phi_m \\ &+ \frac{\alpha n}{\sqrt{2}} \exp(-2i\lambda_a n t/\hbar), \qquad (4) \end{aligned}$$

where γ_m denotes the decay rate of a single molecule imbedded in the atomic condensate: $\gamma_m/\hbar = c_{ma} n$. Under a sudden change of the detuning, from $\epsilon = \epsilon_{in}$ to $\epsilon = \epsilon_f$ at $t = 0$, the molecular field, initially at $\phi_m(t = 0) = \phi_0$, evolves as

$$\begin{aligned} \phi_m(t) = {}& \phi_\infty \exp\!\left[-\frac{it}{\hbar} (2\lambda_a n) \right] + [\phi_0 - \phi_\infty] \\ &\times \exp\!\left[-\frac{it}{\hbar} (\epsilon_f + \lambda n) \right] \exp\!\left(-\frac{\gamma_m t}{2\hbar} \right), \qquad (5) \end{aligned}$$

where ϕ_∞ is the molecular field value at large times, $t \gg (\hbar/\gamma_m)$, $\phi_\infty = -\alpha n/[\sqrt{2}(\epsilon_f + \lambda n - 2\lambda_a n - i\gamma_m/2)] \approx -\alpha n/(\sqrt{2}\, \epsilon_f)$. The molecular population oscillates with angular frequency $\epsilon_f + \lambda n - 2\lambda_a n \approx \epsilon_f$, and magnitude $2|\phi_\infty(\phi_0 - \phi_\infty)| \exp(-\gamma_m t/2\hbar)$, damped out on the scale of the single molecule lifetime. At a later time, the system has relaxed to its "quasiequilibrium" and the molecular field takes on its stationary value, ϕ_∞. For more general off-resonant detuning variations, populations oscillate if $|\dot\epsilon/\epsilon| \ll \gamma_m/\hbar$, or, equivalently, if $|\dot B/(B - B_m)| \ll \gamma_m/\hbar$. In the opposite limit, $|\dot\epsilon/\epsilon| \ll \gamma_m/\hbar$, the system follows its equilibrium value adiabatically: $\phi_m(t) \approx -\alpha n(t)/\sqrt{2}\, \epsilon(t)$. For near-resonant detuning changes [15], the qualitative picture remains: damped population oscillations appear, while molecular decay depletes the condensate, as illustrated in Fig. 1. Experimentally, the oscillations would show, for example, in a modulation of the image intensity obtained with light that is near resonant with the molecules. Contrary to Rabi oscillations of atomic populations, the atom/molecule populations are a pure long-range coherence effect. Even if the relative velocity of the colliding atoms were so low that the atom-molecule populations could oscillate in an individual collision (before the molecule breaks up into atoms that move apart), such oscillations set in when the atoms "meet." In a picture that describes the atoms as classically moving particles, these encounters occur at random times, and any oscillations wash out, as we expect from an incoherent process.

We assume that boson decay is negligible on the time scale of observation, and that the system has reached its ground state [16]. The coherent atom-molecule coupling energy, $\langle \hat H_{\text{at-mol}} \rangle = (\alpha/\sqrt{2}) \int d^3r\, \{\phi_m^* \phi_a^2 + \text{c.c.}\}$,

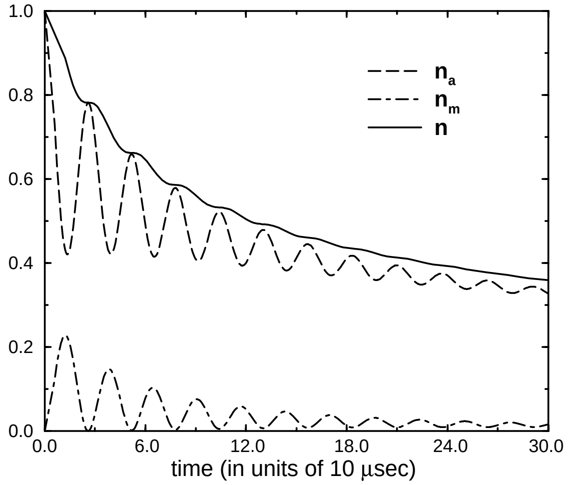

FIG. 1. Plot of the particle densities: the total condensate density, $n = n_a + 2n_m$, is shown in the full line, the atomic density n_a in the dashed line, and the molecular density n_m in the dash-dotted line. The calculation is for a homogeneous BEC that was initially in equilibrium at density $n = 10^{14}$ cm^{-3} when the detuning experienced a sudden shift from $\epsilon = 50\lambda n$ to $\epsilon = 2\lambda n$. The interaction parameters, $\lambda n = \lambda_m n = \lambda_a n = \alpha\sqrt{n} = 10^5$ Hz, and collision rates, $c_{ma} = c_{mm} = 5 \times 10^{-10}$ cm^3 s^{-1} are realistic.

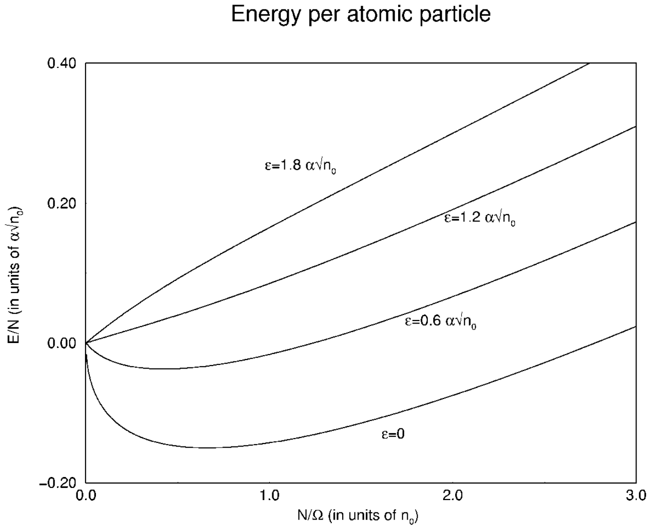

FIG. 2. Ground state energy per atomic particle as a function of the density at different detunings, ϵ. The curves were calculated using realistic values: a reference density $n_0 \sim 10^{14}$ cm^{-3}, and interaction strengths $\lambda_a = \alpha/\sqrt{n_0}$, $\lambda_m = 2\alpha/\sqrt{n_0}$, and $\lambda = 0.2\alpha/\sqrt{n_0}$. The densities at which the minima occur for the two curves of lowest detuning are the self-determined densities that a "free" condensate would adopt in the ground state.

couples the relative condensate phases, so that the lowest energy is reached for a specific phase difference: $\phi_m < 0$ if we choose $\phi_a > 0$. The energy per atomic particle, $e = E/N = \langle \hat{H} \rangle / N$, of a homogeneous BEC, confined to a volume Ω, is then

$$e = \frac{N}{\Omega}\left\{\frac{\lambda_a}{2} f_a^2 + \frac{\lambda_m}{2} f_m^2 + \lambda f_m f_a\right\} - \sqrt{\frac{N}{\Omega}}\{\alpha\sqrt{2f_m}\, f_a\} + \epsilon f_m\,, \tag{6}$$

where f_a (f_m) denotes the fraction of atoms (molecules), $f_a = \Omega n_a/N$, ($f_m = \Omega n_m/N$). Since $f_a = 1 - 2f_m$, the energy is a function of the molecule fraction, the ground state value of which is determined by minimizing e.

In the ultradilute limit, $\Omega \gg N(\lambda_r/\alpha)^2$, where λ_r is representative of the interaction strengths (λ_a, λ_m and λ), the interaction energy $\sim\lambda_r(N/\Omega)$ can be neglected. Close to resonance, $|\epsilon| \ll \alpha\sqrt{(N/\Omega)}$, the detuning is also negligible and the molecule fraction that minimizes the energy is $f_m \approx 1/6$. However, the pressure of the on-resonant ultradilute system, $P = -\partial E/\partial\Omega = -\alpha\sqrt{n}\, n/(3\sqrt{3})$, is negative and the system is mechanically unstable. Interestingly, this instability *does not necessarily imply collapse*. As the system responds to the negative pressure by decreasing its volume, the interaction energy ($\sim\Omega^{-1}$), growing faster than the coupling energy ($\sim\Omega^{-1/2}$), can stabilize the system. In Fig. 2, we show the energy as a function of the atomic particle density. For near-resonant detuning, the tunneling energy dominates in the ultradilute limit, causing the energy to decrease with increasing density. In the high density limit, $\Omega \ll N(\lambda_r/\alpha)^2$, the interaction energy dominates and, for the interaction strengths of Fig. 2, the energy increases linearly with density. In between, the energy reaches a minimum in the density region $\sim(\alpha/\lambda_r)^2$, where interaction and tunneling energies compete. The minimum suggests that the unconfined physical system relaxes to its lowest energy state by adjusting its volume to take on the density of minimum energy. Such self-determined density is a typical liquidlike property, but the calculated values for scattering lengths and α parameter (e.g., for the observed Na resonances) give self-determined densities $\sim 10^{14}$–10^{15} cm^{-3}. The low value is interesting, as the Feshbach resonance could give the first example of a rarified liquid. It is also important, as the condensate could survive spin flip and recombination long enough for the system to be studied.

A direct observation of the liquidlike behavior suggests itself: after switching off the trapping potential, the liquefied condensate remains as a droplet. In a trap, the liquefied condensate takes on a distinct shape, which we now determine for a large condensate in a shallow trap. In a Thomas-Fermi description the local density follows from equating a local chemical potential $\mu(n, \epsilon)$ to the difference of the system's chemical potential, μ_T, and its potential energy. Furthermore, not all atomic particles experience the same external potential: we assume an optical trap in which the atoms experience a trapping potential $V(\mathbf{r})$, but the molecules do not. Alternatively, we may pretend that all atomic particles experience the $V(\mathbf{r})$ potential, as long as we subtract the molecule contribution by assigning a local detuning $\epsilon - 2V(\mathbf{r})$ to the molecules. The Thomas-Fermi equation then reads

$$\mu(n;\epsilon - 2V) = \mu_T - V\,. \tag{7}$$

At the edge, the density abruptly decreases from its self-determined value, n_s, to zero. Around the energy minimum of the homogeneous system,

$$e \approx e_s + \frac{1}{2\kappa n_s}\left(\frac{n - n_s}{n_s}\right)^2. \tag{8}$$

where the self-determined density n_s, the minimum energy e_s, and the compressibility κ ($\kappa^{-1} = -\Omega\,\partial P/\partial\Omega = n_s^3\partial^2 e/\partial n^2$) all depend on the detuning. The chemical potential $\mu(n;\epsilon)$, corresponding to Eq. (8), $\mu = \partial E/\partial N = e + n\,\partial e/\partial n$, is

$$\mu = e_s + \frac{1}{\kappa n_s}\left(\frac{n - n_s}{n_s}\right) + \frac{3}{2\kappa n_s}\left(\frac{n - n_s}{n_s}\right)^2. \tag{9}$$

Finally, in a shallow trap of potential variation significantly less than $\alpha\sqrt{n_s}$, the spatial variation of the detuning may be neglected, $\epsilon - 2V \approx \epsilon$, and Eq. (7) gives

$$n(\mathbf{r}) = \frac{n_s}{3}\left[2 + \sqrt{1 + 6\kappa n_s[V_m - V(\mathbf{r})]}\right], \tag{10}$$

where V_m denotes the potential at the equipotential surface of the boundary. The trap compresses the middle of the droplet causing a density increase that is determined by the compressibility. We estimate the magnitude of the on-resonant compressibility as $\kappa \sim (\lambda_r n_s^2)^{-1}$. Thus, if the density and size of the liquefied atom/molecule condensate is of the order of typical atomic condensates ($n_s \sim 10^{14}$ cm^{-3}, $V_m \sim \lambda_r n_s$), then the trap increases the relative density (n/n_s) by a factor of order 1 (e.g., from 10^{14} cm^{-3} at the edge to 2×10^{14} cm^{-3}, in the middle).

In summary, we have pointed out that the ground state of a Feshbach resonant BEC is a hybrid atomic/molecular condensate. The atomic and molecular condensates interact by coherently exchanging pairs of atoms, and we have discussed implications of this manifestation of quantum coherence: Josephson-like population oscillations in response to a sudden change of the detuning, and the near-resonant liquefaction of the condensate ground state.

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[1] M. H. Anderson *et al.,* Science **269**, 198 (1995).

[2] K. B. Davis *et al.,* Phys. Rev. Lett. **75**, 3969 (1995).

[3] C. C. Bradley *et al.,* Phys. Rev. Lett. **75**, 1687 (1995); **78**, 985 (1997).

[4] They are superfluid in the sense that they are characterized by a complex field, the phase of which determines a superflow.

[5] In addition to the Feshbach resonance, other schemes have been proposed, e.g., by means of external electric fields [see P. W. Milonni and A. Smith, Phys. Rev. A **53**, 3484 (1996); P. O. Fedichev *et al.,* Phys. Rev. Lett. **77**, 2913 (1996)].

[6] These experiments can elucidate fundamental issues as the mechanism of collapse [see, e.g., C. A. Sackett *et al.,* Phys. Rev. Lett. **80**, 2031 (1998); M. Ueda and A. J. Leggett, Phys. Rev. Lett. **80**, 1576 (1998); Yu. Kagan *et al.,* Phys. Rev. Lett. **76**, 2670 (1996); E. Shuryak, Phys. Rev. A **54**, 3151 (1996); M. Ueda and K. Huang, cond-mat/98076359 (to be published)].

[7] W. Stwalley, Phys. Rev. Lett. **37**, 1628 (1976); E. Tiesinga *et al.,* Phys. Rev. A **46**, R1167 (1992); E. Tiesinga *et al.,* Phys. Rev. A **47**, 4114 (1993); A. J. Moerdijk *et al.,* Phys. Rev. A **51**, 4852 (1995); J. M. Vogels *et al.,* Phys. Rev. A **56**, R1067 (1997); H. M. J. M. Boesten *et al.,* Phys. Rev. A **54**, R3726 (1996); J. P. Burke and J. L. Bohn (to be published).

[8] S. Inouye *et al.,* Nature (London) **392**, 151 (1998); Ph. Courteille *et al.,* Phys. Rev. Lett. **81**, 69 (1998); J. L. Roberts *et al.,* Phys. Rev. Lett. **81**, 5109 (1998); V. Vuletic *et al.,* Phys. Rev. Lett. **82**, 1406 (1999).

[9] This procedure was first developed in P. Tommasini *et al.,* cond-mat/9804015 (to be published); E. Timmermans *et al.,* cond-mat/9805323 (to be published); R. Côté *et al.,* in Proceedings of E.U. meeting on New Directions in Atomic Physics, edited by C. T. Whelan *et al.* (Plenum, New York, to be published); E. Timmermans *et al.,* Phys. Rep. **315**, 199 (1999). The ideas proposed in these papers were followed up by Piza *et al.* [(to be published)], and by P. D. Drummond *et al.* [Phys. Rev. Lett. **81**, 3055 (1998)].

[10] The interaction strengths λ' depend on the scattering lengths a' and the reduced mass m'_{red} for the corresponding collision: $\lambda' = 2\pi\hbar^2 a'/m'_{\text{red}}$.

[11] E. P. Gross, Nuovo Cimento **20**, 454 (1961).

[12] L. P. Pitaevskii, Sov. Phys. JETP **13**, 451 (1961).

[13] Alternatively, within the framework of the Gaussian approximation, one obtains the equations of motion from an effective Hamiltonian [see A. Kerman and P. Tommasini, Ann. Phys. (N.Y.) **260**, 250 (1997)].

[14] These estimates follow from calculations on hydrogen and helium, recently reported in N. Balakrishnan *et al.,* Phys. Rev. Lett. **80**, 3224 (1998).

[15] At arbitrary detuning, the oscillation frequency can be derived from a Hamiltonian description of the population dynamics [see Phys. Rep. of Ref. [9]].

[16] Alternatively, one may solve for imaginary eigenvalues of the equations of motion with imaginary interaction strengths.

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