Dynamics of a molecular Bose-Einstein condensate near a Feshbach resonance

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We consider the dissociation of a molecular Bose-Einstein condensate due to a magnetic-field sweep that starts on the molecular side of a Feshbach resonance and ends either on the atomic or on the molecular side. We determine both the time-dependent response of the molecular condensate wave function and the energy distribution of the atoms produced after the sweep. In the cases where experiments have been performed, our analytic and numerical calculations provide good agreement with experimental atomic energy distributions.

I. INTRODUCTION

Feshbach resonances are now one of the most important tools in the area of ultracold atomic gases [1]. These resonances provide the opportunity to change with an applied magnetic field the energy of a diatomic molecular state from a region where the molecules are more stable to a region where the atoms are more stable. As a result, sweeping an ultracold gas across or near a resonance is at present the most important application of Feshbach resonances. A dramatic early experiment observed in this manner Josephson oscillations between atomic and molecular condensates [2]. More recently, a variety of sweeps have been employed to create an ultracold molecular gas starting from a degenerate fermionic atom gas [3,4] and vice versa [5], or to obtain evidence for a fermionic condensate [6]. In addition, two experiments have employed magnetic-field sweeps to drive ultracold molecular gases across the Feshbach resonance to produce bosonic atoms with a nonequilibrium energy distribution [7,8].

Theoretical treatments of sweeps from the molecular to the atomic side have been restricted to long sweeps with constant sweep rates, and have assumed the sweep to be almost adiabatic [7,9]. The assumption of adiabaticity is difficult to justify in the case of a uniform gas, where there is a continuum of atomic levels rather than a discrete one, so that it is difficult to define "slow" sweeps with respect to an energy gap in the spectrum of adiabatic states. In this article, we provide a procedure for analyzing sweeps starting from a molecular condensate that is valid for essentially arbitrary sweep forms and is free of assumptions of adiabaticity. We show that under appropriate conditions the statistics of the produced atoms can be neglected, and in that case our results are equally valid for fermionic and bosonic atoms.

We focus in particular on two quantities. First, we develop a method for determining the decay of the molecular condensate during the sweep. Second, we investigate for different kinds of sweeps the resulting energy spectrum of the dissociated atoms, which turns out to be closely related to the time evolution of the molecular condensate. Our approach to the time evolution is based on the equation of motion for the molecular condensate wave function $\phi_m(t)$, developed by Duine and Stoof [1,10]. While no experiment, to our knowledge, has yet probed the molecular dissociation as a function

of time, the decay of the molecular condensate density is in principle experimentally accessible by a series of destructive measurements. For two classes of sweeps, we also provide exact analytic expressions for both the time-dependent wave function $\phi_m(t)$ and the energy distribution of dissociated atoms.

Measurements of atomic energies after a dissociation ramp have been performed recently by Mukaiyama *et al.* [7] and by Dürr and co-workers [8]. The ramps of Ref. [7] are restricted to cases where all the molecules dissociate during the sweep. The shape of the dissociation spectrum for this type of sweep is well described by one of our exact analytical results. Only a special case of this result was available previously [7,9]. In Ref. [8] also a different, more rapid type of sweep experiment is reported, in which most of the dissociation occurs *after* the sweep is over. As a result there is now a sharp peak in the atomic energy distribution corresponding to half the molecular energy at the end of the sweep. Our approach also allows us to calculate $\phi_m(t)$ and the atomic spectra for such fast sweeps. For the extreme case of an instantaneous sweep, we also have analytic results. In addition, we also consider a new type of sweep, one that keeps the detuning negative while bringing it closer to the resonance. This kind of sweep results in several interesting physical effects: a rearrangement of the dressed molecule structure, incoherent decay, and Josephson oscillations between molecular and atomic condensate components.

II. GENERALIZED GROSS-PITAEVSKII EQUATION

A Feshbach resonance is characterized by its magnetic field location B_0, width ΔB, background scattering length a_{bg}, and magnetic-moment difference $\Delta\mu$ between two atoms and the bare molecule associated with the Feshbach resonance. Assuming that these quantities are for the atomic gas of interest known from experiment, the equation of motion for the bare molecular condensate wave function $\phi_m(t)$ becomes [1]

$$i\hbar\partial_t\phi_m(t) = [\delta(t) - i\eta\sqrt{i\hbar\partial_t}]\phi_m(t), \qquad (1)$$

where $\delta(t) = \Delta\mu[B(t) - B_0]$ is the so-called detuning that is time dependent during the sweep. The imaginary term in this equation of motion describes the decay of molecules into

pairs of atoms and is determined in the Bose case by the quantity $\eta = m^{1/2} a_{bg} \Delta B \Delta \mu / \hbar = m^{3/2} g^2 / 2\pi\hbar^3$, where g is the atom-molecule coupling. When the atoms are fermionic, the right-hand side is a factor of 2 smaller. Note that η^2 has the units of energy and physically quantifies the width of the resonance. The imaginary term arises from the molecular self-energy

$$\hbar\Sigma_m(z) = \quad\Longrightarrow\bigcirc\Longrightarrow\quad = \eta\sqrt{-z}, \qquad (2)$$

where the single and double lines represent atomic and molecular propagators, respectively. The square root of energy reflects the three-dimensional final density of states expected from a Fermi's Golden Rule calculation of the dissociation process. The $\sqrt{i\hbar\partial_t}$ form in Eq. (1) arises from evaluating the complex self-energy in Eq. (2) from above on the real axis. The nonlocality of this operator in time implies history dependence in the dynamics.

The above self-energy leads physically to a dressing of the molecules. The resulting molecular density of states contains, for negative detuning, a delta function with weight $Z = (1 + \eta/2\sqrt{|\epsilon_m|})^{-1}$ at the dressed molecular binding energy $\epsilon_m(\delta) = \delta - \eta^2/2 + \eta\sqrt{\eta^2/4 - \delta}$ [1]. The ϕ_m appearing in Eq. (1) represents the condensate wave function for bare molecules. The dressed molecular density is a factor Z^{-1} larger than the bare density and equal to $|\phi_m|^2/Z$. Moreover, the bare molecular density can now change in two ways. Coherent changes involve a shift in the linear superposition making up the dressed molecular wave function, i.e., a change of the bare molecular fraction Z, while the dressed molecular density remains constant [1,2]. Incoherent changes are associated with the breakup of the dressed molecule itself.

The generalized Gross-Pitaevskii equation in Eq. (1) involves some approximations. First, we are considering sufficiently low molecular densities that the molecule-molecule interactions can be neglected for most of the sweep duration. Second, we ignore the thermal part of the molecular gas, which is justified for sufficiently low temperatures. Most importantly, we are neglecting feedback effects, i.e., the dynamics and statistics of the atoms that are formed during and after the sweep. In particular, we do not allow for their reassociation. The neglect of atomic statistics, i.e., Bose enhancement or Pauli blocking, is reasonable for monotonic sweeps, because the dissociation then always occurs at different energies. In that case we can estimate that the atomic statistics can be absolutely neglected if the molecular density obeys $n_m < \hbar\dot{\delta}/\sqrt{2}\pi^2 g^2$.

III. NUMERICAL METHOD

We now outline the method of solving Eq. (1), given a particular time-dependent detuning $\delta(t)$. We are especially interested in magnetic-field sweeps across the resonance that take the detuning δ from a negative value δ_i at $t=0$ to the final positive or negative value δ_f at time $t=T$. Introducing an auxiliary constant detuning δ_c, we can write

$$(i\hbar\partial_t + i\eta\sqrt{i\hbar\partial_t} - \delta_c)\phi_m(t) = [\delta(t) - \delta_c]\phi_m(t). \qquad (3)$$

The solution of this differential equation is the sum of a particular solution and the solution of the corresponding homogeneous equation. We thus have

$$\phi_m(t) = \psi(t) + \int_{-\infty}^{t} dt' G_c(t-t')[\delta(t') - \delta_c]\phi_m(t'). \qquad (4)$$

Here $G_c(t)$ is the Green's function for the operator acting on $\phi_m(t)$ in the left-hand side of Eq. (3), i.e.,

$$G_c(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{e^{-i\omega t}}{\hbar\omega^+ - \eta\sqrt{-\hbar\omega^+} - \delta_c}, \qquad (5)$$

where $\hbar\omega^\pm = \hbar\omega \pm i0$. The real part of the pole in $G_c(\omega)$ gives the molecular energy and the imaginary part gives the dissociation rate, both at detuning δ_c. Moreover, $\psi(t)$ is the solution of the homogeneous equation of Eq. (3). For $\delta_c < 0$, the solution is an oscillatory function, $\psi(t) \propto e^{-i\omega_c t}$, with a frequency given by $\hbar\omega_c = \epsilon_m(\delta_c)$. It is convenient to use $\delta_c = \delta_i$, because the integral in Eq. (4) then has a lower limit of 0 instead of $-\infty$.

The retarded Green's function $G_c(t)$ is calculated by closing the contour in the negative half plane. The integrand has a branch cut because of the square root in the energy denominator. For $\delta_c < 0$, there is also a pole at $\epsilon_m(\delta_c)$. Taking these into account we obtain

$$G_c(t) = \theta(t)\left[-\frac{i\eta}{\pi}\int_0^{\infty} d\omega \frac{\sqrt{\hbar\omega}e^{-i\omega t}}{(\hbar\omega - \delta_c)^2 + \eta^2\hbar\omega} \right.$$
$$\left. -\frac{i}{\hbar}e^{-i\omega_c t}\frac{2\sqrt{-\hbar\omega_c}}{\eta + 2\sqrt{-\hbar\omega_c}} \right].$$

Having calculated the Green's function $G_c(t)$ and the wave function $\psi(t)$ corresponding to δ_c, we can numerically solve Eq. (4) for $\phi_m(t)$.

IV. EVOLUTION OF CONDENSATE WAVE FUNCTION

In Fig. 1 we show the numerically calculated decay of the bare molecular wave function $\phi_m(t)$ for two different ramps, shown in the top panels. The center panels show the oscillatory decay of the real part of $\phi_m(t)$. For the constant-rate or "long" sweep shown in the left column, we see that $\phi_m(t)$ has a gradually decreasing oscillation frequency, corresponding to the decreasing magnitude of the binding energy $\epsilon_m(\delta)$ as δ approaches zero from the negative side. The decay of the bare density $n_m(t) = |\phi_m(t)|^2$ shown in the bottom panel is mostly coherent for $\delta(t) < 0$, so that the dressed molecule density remains almost fixed. On the other hand, the decay at positive detuning is incoherent, and the decay of the bare density $n_m(t)$ corresponds to the decay of dressed molecules. The ramp in the experiment of Mukaiyama et al. [7] involves an initial and final detuning $\delta_{i,f}$ of about $\mp 50\eta^2$ and ramp times of $T \gtrsim 100\hbar/\eta^2$. For the results shown in the left column of Fig. 1, we used smaller values for both so as to have fewer oscillations to visualize. For the instantaneous sweep
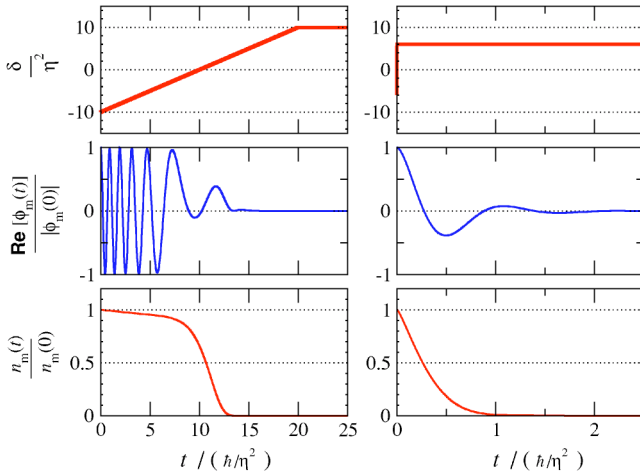
FIG. 1. (Color online) Decay of the molecular wave function $\phi_m(t)/|\phi_m(0)|$ with time. Left column: detuning changes linearly with time from $\delta_i = -10\eta^2$ to $\delta_f = +10\eta^2$. Right column: instantaneous sweep from $\delta_i = -6\eta^2$ to $\delta_f = +6\eta^2$. Top panels show the sweeps. Center panels show real part of $\phi_m(t)$. Bottom panels show the decay of the molecular density, $n_m(t)/n_m(0) = |\phi_m(t)|^2/|\phi_m(0)|^2$.

of the right column, we see $\phi_m(t)$ undergoing oscillatory decay after the ramp. For $\delta_f \gg \eta^2$, the scale of the decay and the oscillations are of order $\hbar/(\eta\sqrt{\delta_f})$ and $\hbar/\delta_f$, respectively. However, the detailed evolution depends on the history, i.e., on δ_i.

For two special but experimentally relevant types of sweeps, we can also provide analytic solutions for Eq. (1). The first case is when the ramp speed is constant, and it continues after all the molecules are dissociated. The solution is then given by the generalized Fourier transform

$$\phi_m(\omega) = B \exp\left[i\frac{\hbar\omega^2}{2\dot\delta} - i\frac{\delta_i\omega}{\dot\delta} + \frac{2\eta}{3\hbar\dot\delta}i(-\hbar\omega)^{3/2} \right], \quad (6)$$

with $\phi_m(t) = \int_C (d\omega/2\pi)\phi_m(\omega)e^{-i\omega t}$ and $|B|^2 = 2\pi\hbar n_m(0)/Z_i\dot\delta$. The contour first runs through the third quadrant to ensure convergence and then runs above the branch cut along the positive real axis. The second case is that of ultrafast ramps, when the detuning is switched from negative $\delta(t) = \delta_i$ to positive $\delta(t) = \delta_f$ instantaneously at $t=0$. The solution in this case is

$$\phi_m(t) = (\delta_f - \delta_i)\sqrt{n_m(0)}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\frac{\hbar G_f(\omega)e^{-i\omega t}}{\hbar\omega^- - \epsilon_m(\delta_i)}. \quad (7)$$

Here G_f is the Green's function corresponding to $\delta = \delta_f$. The analytic expressions in Eqs. (6) and (7) for $\phi_m(t)$ reproduce the numerical calculations for sweeps of the applicable types, such as those in the left and right panels of Fig. 1, respectively.

V. ATOMIC ENERGY DISTRIBUTION

We now outline how the incoherent dissociation spectrum is obtained once $\phi_m(t)$ has been calculated. We calculate the time derivative of the molecular density from the equation of motion in Eq. (1) and find
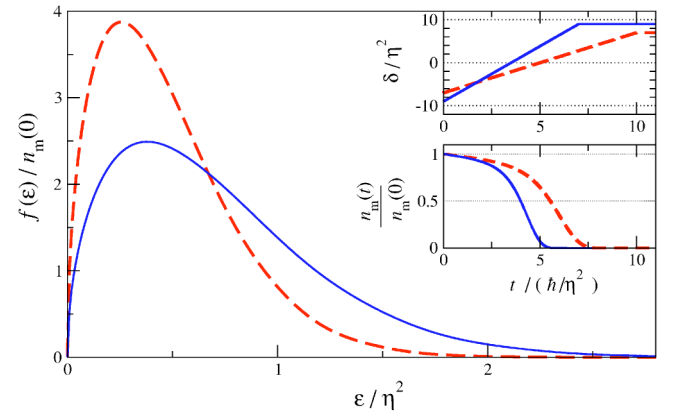

FIG. 2. (Color online) Spectra for constant-rate "long" sweeps, where the molecules decay almost completely before the sweep is over. Insets show the detuning ramps with different ramp rates and the bare molecular condensate densities.

$$\int_{-\infty}^{\infty} dt\, \dot n_m\bigg|_{\text{incoh}} = -\frac{\eta}{\pi\hbar}\int_0^{\infty} d\omega\sqrt{\hbar\omega}|\phi_m(\omega)|^2. \quad (8)$$

The integrand in the right-hand side is interpreted as the molecular dissociation spectrum $f_{\text{mol}}(\epsilon) = \eta\sqrt{\epsilon}|\phi_m(\epsilon/\hbar)|^2/\pi\hbar^2$. Since each atom has half the energy of the dissociating molecule, $f_{\text{mol}}(\epsilon)$ corresponds to the atomic spectrum at $\epsilon/2$, i.e., $f_{\text{at}}(\epsilon) = 4f_{\text{mol}}(2\epsilon)$.

For constant-rate "long" sweeps where all molecules decay before the sweep is over, our analytic result for $\phi_m(\omega)$ yields for the atomic spectrum

$$f_{\text{at}}(\epsilon) = Z_i^{-1}n_m(0)\frac{8\eta\sqrt{2\epsilon}}{\hbar\dot\delta}\exp\left[-\frac{4\eta(2\epsilon)^{3/2}}{3\hbar\dot\delta}\right]. \quad (9)$$

The factor $Z_i^{-1} = [Z(\delta_i)]^{-1}$ represents the fact that it is the dressed molecule density, rather than the bare molecule density, that determines the final atomic density. Without the normalization factor Z_i^{-1} due to the initial dressing of the molecules, the expression in Eq. (9) has been derived before from Landau-Zener [9] and from rate-equation [7] considerations. The normalization is unimportant only for $|\delta_i| \gg \eta^2$, in which case $Z_i \simeq 1$.

In Fig. 2 we show numerically calculated spectra for two "long" sweeps. The spectra match the exponential form of Eq. (9). The Z_i^{-1} factor is important in these cases. The sweep represented by the solid curves has a higher ramp rate and hence the resulting spectrum is tilted to higher energies. Note that experimentally only the total atomic energy has been measured as a function of ramp speeds [7,8]. These measured total energies are fit well by the total energies calculated analytically as $\int d\epsilon\,\epsilon f_{\text{at}}(\epsilon)$. Since our spectra match the analytic result for the experimentally relevant case of $Z_i \simeq 1$, agreement with the measured total energies follows.

In Fig. 3 we show spectra calculated for two cases where Eq. (9) does not hold because the sweep is over before all the molecules have decayed. This type of sweep has been employed in a recent experiment [8]. The density images in that experiment indicate that the spectrum is peaked at the energy
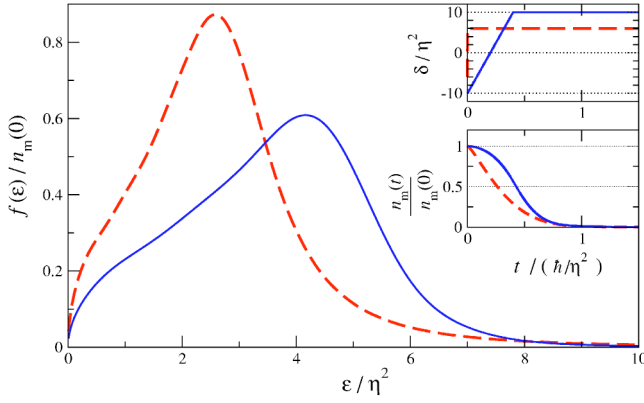
FIG. 3. (Color online) Spectra for fast and short sweeps. Dashed curves: instantaneous ramp in which the detuning jumps from $\delta_i = -6\eta^2$ to $\delta_f = +6\eta^2$ at $t=0$. Full curves: fast ramp that is over before all the molecules can decay.

corresponding to the final detuning. This feature is clear in Fig. 3. The dashed curves represent an infinitely fast ramp. The spectrum shows a peak around $\delta_f/2$, but is spread over a relatively large range of energies. The spread demonstrates that our method takes into account the distribution of molecular energies, i.e., the width of the molecular density of states at a fixed positive detuning. For the instantaneous ramp, our exact solution for $\phi_m(t)$ in Eq. (7) leads to the analytic result

$$f_{at}(\epsilon) = \frac{4\eta}{\pi}\left(\frac{\delta_f - \delta_i}{2\epsilon - \hbar\omega_i}\right)^2 \frac{n_m(0)\sqrt{2\epsilon}}{(2\epsilon - \delta_f)^2 + 2\eta^2\epsilon}, \qquad (10)$$

which describes exactly the dashed spectrum of Fig. 3. The solid curves in Fig. 3 represent a ramp that is over before all the molecules have decayed. A significant fraction decays while the detuning is constant at its final value. The atomic spectrum here also shows a peak at an energy around half the final detuning. No analytic expressions are available for this kind of sweep.

In order to demonstrate mainly coherent loss of bare molecules, we display in Fig. 4 a sweep that ends at negative detuning, i.e., before reaching the resonance. The density of bare molecules decreases, but most of this decrease is due to the coherent change of the dressed molecular wave function. The final density n_f is slightly smaller than $(Z_f/Z_i)n_m(0)$, where Z_i and Z_f are the weights of the delta function in the molecular density of states at the initial and final detunings, respectively. The difference between n_f and $(Z_f/Z_i)n_m(0)$ appears as the total weight of the incoherent spectrum. Note also that the density undergoes some coherent oscillations before settling at its final value. These are internal Josephson oscillations of the dressed molecular condensate.

VI. CONCLUSIONS

We have shown how to calculate the time evolution of the molecular condensate wave function $\phi_m(t)$ during sweeps
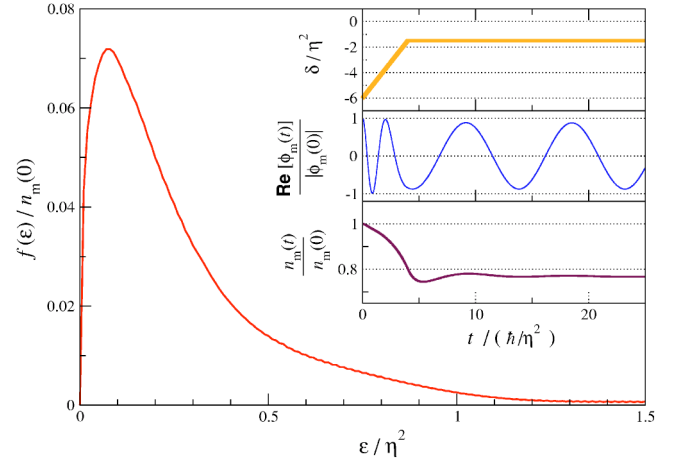

FIG. 4. (Color online) A sweep that starts and ends at negative detuning. In this case, the final density is expected to be $(Z_f/Z_i)n_m(0) \simeq 0.7775 n_m(0)$. The actual final density, $n_f \simeq 0.735 n_m(0)$, is smaller, in agreement with the nonzero atomic spectrum.

across and near a Feshbach resonance. We have also shown how the time dependence of this wave function leads to the dissociation spectrum. The theory presented here is applicable for ramps of essentially arbitrary shape and initial conditions. We have also obtained quantitative analytic expressions for some cases, both for the time evolution and for the atomic spectrum. These analytic results are more general than the special case treated in Refs. [7,9]. Moreover, we have obtained the, in first instance surprising, result that a nonzero dissociation spectrum can also be obtained by a changing magnetic field that always remains below the Feshbach resonance.

Finally, we point out two issues that remain unsolved. First is the problem of incorporating the effect of the dynamics and quantum statistics of the atoms. In principle, this can be achieved by coupling the generalized Gross-Pitaevskii equation for the molecular condensate to a quantum Boltzmann equation for the atoms. It remains to be seen, however, if this leads to a feasible approach. Second, our calculation is based on the approximation for the molecular self-energy given in Eq. (2). For some broad Feshbach resonances, such as the one in ^{6}Li, the molecular self-energy depends not only on η but also on the background scattering length a_{bg} [1]. Incorporating this improvement is left for future work.

[1] R. A. Duine and H. T. C. Stoof, Phys. Rep. **396**, 115 (2004).

[2] E. A. Donley, N. R. Claussen, S. T. Thompson, and C. E. Wieman, Nature (London) **417**, 529 (2002).

[3] K. E. Strecker, G. B. Partridge, and R. G. Hulet, Phys. Rev. Lett. **91**, 080406 (2003).

[4] M. Greiner, C. A. Regal, and D. S. Jin, Nature (London) **426**, 537 (2003).

[5] M. Bartenstein, A. Altmeyer, S. Riedl, S. Jochim, C. Chin, J. H. Denschlag, and R. Grimm, Phys. Rev. Lett. **92**, 120401 (2004); **92**, 203201 (2004).

[6] C. A. Regal, M. Greiner, and D. S. Jin, Phys. Rev. Lett. **92**, 040403 (2004); M. W. Zwierlein, C. A. Stan, C. H. Schunck, S. M. F. Raupach, A. J. Kerman, and W. Ketterle, *ibid.* **92**, 120403 (2004).

[7] T. Mukaiyama, J. R. Abo-Shaeer, K. Xu, J. K. Chin, and W. Ketterle, Phys. Rev. Lett. **92**, 180402 (2004).

[8] S. Dürr, T. Volz, and G. Rempe, Phys. Rev. A **70**, 031601(R) (2004).

[9] K. Góral, T. Köhler, S. A. Gardiner, E. Tiesinga, and P. S. Julienne, J. Phys. B **37**, 3457 (2004).

[10] R. A. Duine and H. T. C. Stoof, J. Opt. B: Quantum Semiclassical Opt. **5**, S212 (2003).