Laser-Driven Collisions between Atoms in a Bose-Einstein Condensed Gas

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We have determined the rate of loss of atoms from a Bose-Einstein condensed gas due to binary processes in the presence of a far-detuned laser field. In this limit, the binary loss rate spectrum is markedly different from, and can greatly exceed, the basic atomic loss rate. We suggest that measurements of the loss rate spectrum can be used to determine the nature of atom interactions in a condensate. [S0031-9007(96)00919-2]

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The recent achievement of Bose-Einstein condensation (BEC) in alkali gases using the combination of laser and evaporative cooling have given a new urgency to the study of ultracold atomic and molecular processes [1–3]. Laser cooling alone is not sufficient to reach condensation, due to the atomic recoil heating and collision-induced loss processes that occur in the presence of a laser field. At present the only way to achieve the necessary conditions for BEC (temperatures below 1 μK and densities above 10^{13} atoms/cm^3) is to use evaporative cooling on a gas precooled using laser techniques. Once a condensate is formed, however, one would like to know if light can be used to study its properties. In addition, laser fields may be used for controlling the condensed atoms and for doing atom optics [4]. This will most likely be a major theme in the years ahead as one attempts to use condensed gases for interferometry, probes of the properties of matter, and lithography. One important application has been the use of a far-detuned laser to remove the zero magnetic field hole in a quadrupole trap in the MIT BEC experiment [3], but there also are proposals for achieving BEC using laser-driven schemes [5]. There is, of course, the closely related issue of producing an atom laser: This is the equivalent of a laser for atoms and is a source of coherent matter waves. Again, some schemes for making one rely on the presence of a laser field [6,7].

For these reasons it is important to look at how the presence of a laser field influences the atomic interactions in a condensate. The theory of light interaction with condensates has been discussed by several authors [8–10], and we shall use part of their work in our treatment. The theories given so far do not address in detail the issue of how strong binary interactions will affect the evolution of the condensed gas, and this is what we shall do here. In particular, the condensate will have a photoassociation spectrum [11–16], which we suggest will be a good diagnostic of atomic interactions in the condensate. The extremely low temperature of the evaporatively cooled gases also permits us to give an accurate analytical expression which is much simpler than that for the thermally broadened line shapes [17] in laser cooled and trapped gases.

The first issue we have to address is when one can treat the interactions within a condensate induced by the light field as binary events. Although the condensed atoms form a single quantum system, it is still proper to deal with the strong dipole-dipole interactions involving excited atoms in the gas using the binary collision approach in certain limits. This does not mean that the propagation of light in the medium is a pure binary process. The propagation can be handled using a polarization or a multiple scattering technique. The formalism for doing this has been set up in Ref. [8].

In the dilute gas limit the many-body T matrix approach handles the onset of multiple particle effects by including the occupation of intermediate states in the perturbation expansion [18]. We have to ask, therefore, what is the occupation of the relevant states in a collision in which dipole-dipole interactions are dominant. The only state that can play an appreciable role is, of course, the macroscopically occupied condensate. Strong dipole-dipole interactions, however, give the colliding pair momenta that are very large compared to the small amount that condensed particles have (they do have some because of the trap). This means that the effect of the condensate in the middle of the dipole interactions can be quite safely ignored. This is distinct from the case of the weak elastic ground state interactions where, particularly for the case of attractive interactions, the many-body changes to the T matrix produce profound effects [19]. Ignoring these effects in the binary interactions is only valid, however, if the detunings are sufficiently large, an issue that has been addressed in Ref. [9].

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There are many-body effects that we have to include explicitly, i.e., for cases where we excite close to resonance we have to allow for the fact that for long range interactions the atoms may be able to make transitions back to the condensate. This possibility has in effect two consequences. The first is a collective effect, i.e., the superradiant enhancement of the excited atom decay rate $\gamma_{\rm con}$, which may be shown to have the following form [9]:

$$\gamma_{\rm con} = (n\lambda\!\!\!^{-3}_A)\left(\frac{l}{\lambda\!\!\!^{-}_A}\right)\gamma_A\,, \tag{1}$$

where l is a typical linear dimension of the trap, n is the particle density in the condensate, $\lambda\!\!\!^{-}_A = \lambda_A/2\pi$, where λ_A is the wavelength of the atomic resonance transition at frequency ω_A, and γ_A is the natural atomic decay rate.

We should emphasize that this enhancement of the decay rate and hence the width of the line does not necessarily lead to extra loss from the condensate due to light scattering. This is because the transitions contributing to $\gamma_{\rm con}$ end up back in the condensate. The second effect that we have to consider, in principle, is the change in the binary interaction due to this enhanced decay. To avoid both of these complications we shall suppose we are detuned more than the collective width of the line. This means that we can treat the interaction as being dominantly binary. We can conclude, therefore, that for detunings that are much larger than $\gamma_{\rm con}$ we can treat the light-induced collisions as binary events. In particular, we shall compare the binary rate $\Gamma_{\rm binary}$ for photoinduced loss of atoms from the condensate (and probably from the trap as well) with the loss rate $\Gamma_{\rm atomic}$, due to single atom recoil:

$$\Gamma_{\rm atomic} = \gamma_A\left(\frac{\Omega_A}{\Delta_A}\right)^2, \tag{2}$$

where $\hbar\Omega_A$ is the atomic optical coupling matrix element and $\Delta_A = \omega - \omega_A \gg \gamma_{\rm con}$ is the detuning from the atomic resonance frequency. We now turn to the detailed calculation of $\Gamma_{\rm binary} = nK_{\rm loss}$, where n is the condensate density and $K_{\rm loss}$ is the binary rate coefficient. Although our results are expressed in analytic form for a simple excited state model, they are obtained from a fully quantum mechanical model for ultracold collisions in a weak radiation field [20], justified by quantum scattering calculations, and can be generalized to apply to real alkali atoms with hyperfine structure [21,22].

The effect of binary collisions depends strongly on whether the laser detuning Δ_A is to the red or blue of the atomic resonance ω_A. The resonant dipole interaction gives rise to attractive and repulsive states, as shown in Fig. 1. Blue detuning, therefore, excites to a repulsive state, which dissociates and releases $\sim\hbar\Delta_A$ in kinetic energy shared between the atoms. Red detuning, on the other hand, excites bound levels of the attractive potential. The subsequent decay of such vibrational levels, at molecular rate γ_v, results in the loss of trapped atoms,

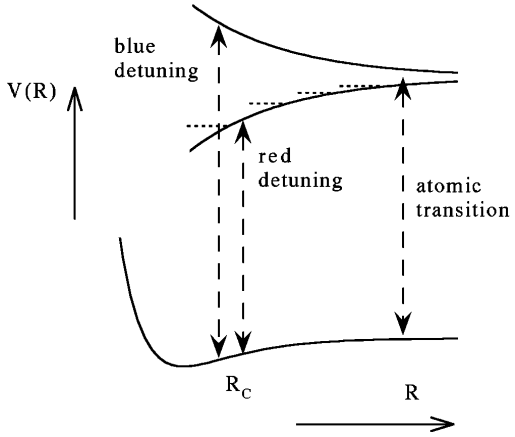

FIG. 1. The typical ground and excited state configuration for cold collisions in light fields in the detuning regime where the excited state structure is dominated by the dipole-dipole interaction. The attractive excited state will become strongly repulsive at small R (not shown), and has thus bound state structure, whereas the repulsive state leads to dissociation.

since most of the fluorescence is to the untrapped states. We shall assume that the light intensity is low enough that the photoinduced collision rates can be calculated perturbatively in the strength of the light field.

At very low T, only s waves contribute to collisions and

$$K_{\rm loss} = 2\left(\frac{2-x}{2}\right)\frac{\pi\hbar}{\mu k}\,\langle|S_{ge}(E,\Delta_A)|^2\rangle\,, \tag{3}$$

where the first factor of 2 results from two atoms lost per collision, and the $\frac{2-x}{2}$ factor is due to the presence of a condensate with fraction x [23]. Here $|S_{ge}|^2$ is the s-wave S-matrix element for the photoinduced loss process, μ is the reduced mass for the colliding pair, $\hbar k = (2\mu E)^{1/2}$ is the asymptotic momentum for their relative motion, and the brackets imply an average over the distribution of k.

For blue detuning and weak optical coupling

$$|S_{ge}(E,\Delta_A)|^2 = 4\pi^2|\langle\Psi_g(E)|V_{ge}|\Psi_e(E+\hbar\Delta_A)\rangle|^2. \tag{4}$$

Here Ψ_g and Ψ_e are the energy-normalized ground and excited state wave functions, respectively, and $V_{ge} = \hbar\Omega(R) = \hbar b(R)\Omega_A$ is the optical coupling matrix element. The function $b(R)$ relates the molecular optical coupling to the atomic. For red detunings, the collision matrix element depends sensitively on the positions of the molecular bound states [17]:

$$|S_{ge}(E,\Delta_A)|^2 = \sum_v \frac{\gamma_v\,\gamma_s(E,v)}{(E/\hbar+\Delta_A-\Delta_v)^2+(\gamma_v/2)^2}\,, \tag{5}$$

where

$$\gamma_s(E,v) = \frac{2\pi}{\hbar}\,|\langle\Psi_g(E)|V_{ge}|\Psi_v\rangle|^2. \tag{6}$$

Here Ψ_v is the wave function corresponding to the vibrational level v of the bound excited state, and Δ_v is its position (detuned from the atomic resonance). In conventional photoassociation spectroscopy [11–16], the spread of E is comparable to $\gamma_v/2$, and the variation with E of both the numerator and denominator in Eq. (5) affects the photoassociation probability, requiring numerical evaluation of the thermal line shapes. The spread of E is so small for evaporatively cooled gases that E can be set to zero in the denominator of Eq. (5). In addition, $\gamma_s(E, v) \propto k$ so that $|S_{ge}|^2/k$ is independent of E as $k \to 0$. Thus the brackets in Eq. (3) are unnecessary for this case. The same is true for blue detuning, Eq. (4).

In both cases, for a slowly varying $\Omega(R)$, we can rewrite the squares of the coupling matrix elements as $\Omega(R_C)^2 f_C$, where R_C is the Condon point, i.e., the resonance point for the quasimolecule (as illustrated in Fig. 1) for which $\hbar\Delta_A = V_e(R_C) - V_g(R_C) \approx C_3/R_C^3$ (C_3 is the dipole-dipole potential coefficient and V_g and V_e are the ground and excited state potentials, respectively), and f_C is the Franck-Condon factor between the ground state wave function $\Psi_g(R, E)$ and the excited state wave function; a free state with energy $E + \hbar\Delta_A$ for blue detuning: $f_C = |\langle\Psi_g(E)|\Psi_e(E + \hbar\Delta_A)\rangle|^2$, and a bound state v with binding energy $\hbar\Delta_v = \hbar\Delta_A$ for the case of red detuning: $f_C = (\langle\Psi_g(E)|\Psi_v\rangle)^2$. Since the ground state wave function varies so much more slowly with R than the excited state wave function, a simple reflection approximation provides an accurate expression for the Franck-Condon factors [20]

$$\text{blue: } f_C = \frac{1}{D_C}\,|\Psi_g(R_C, E)|^2, \tag{7}$$

$$\text{red: } f_C = h\nu_v \frac{1}{D_C}\,|\Psi_g(R_C, E)|^2. \tag{8}$$

Here $D_C = |d(V_e - V_g)/dR|$ is the slope of the difference potential at $R = R_C$, and ν_v is the vibration frequency for the excited bound level v. The derivation of the above expressions in Ref. [20] follows the Appendix of Ref. [24], generalized to low collision energies; the $1/D_C$ factor results from the familiar stationary phase approximation for Franck-Condon factors [24].

The Franck-Condon factor f_C is proportional to the square of the ground state wave function at R_C. The spectrum, therefore, serves as a probe of the ground state pair correlation function. This shows that measurements of the optical spectra due to strongly interacting pairs in a condensate might be used to study such correlations, as suggested in Ref. [25].

A simple analytic expression for the ground state wave function at long range, $R > R_B$, corrected for the effect of the long range $-C_6/R^6$ van der Waals potential, is given by [20]

$$\left(\frac{2\mu}{\pi\hbar^2 k}\right)^{1/2} a(R)\sin[k\rho(R)], \tag{9}$$

where

$$a(R) = 1 - \left(\frac{R_B}{R}\right)^4, \quad \rho(R) = R - A_s - \frac{2}{3}\left(\frac{R_B}{R}\right)^4 R\,. \tag{10}$$

Here A_s is the scattering length for the ground state potential, and $R_B = (\mu C_6/10\hbar^2)^{1/4}$ has the values $42a_0$ for Na and $77a_0$ for Rb. If the correction due to the long range potential is neglected, the wave function takes on the simpler form: $a = 1, \rho(R) = R - A_S$ [14].

At the Condon point we write $\Omega(R_C) = b_C\Omega_A$ and use the fact that $C_3 = f_3\hbar\gamma_A\lambda\!\!\!^{-3}_A$, where the molecular structure factors $0 < b_C < 2/\sqrt{3}$ and $0 < |f_3| < 1/2$ depend on the particular molecular states involved. For blue detuning the binary loss rate per atom, $\Gamma^{\text{blue}}_{\text{binary}} = K_{\text{loss}}n$, is

$$\Gamma^{\text{blue}}_{\text{binary}} = \Gamma^{(1)}_{\text{binary}} = (2 - x)\,\frac{8\pi^2 b_C^2 f_3}{3}\,(n\lambda\!\!\!^{-3}_A) g_C \Gamma_{\text{atomic}} \tag{11}$$

when $g_C = [1 - (R_B/R_C)^4]^2[1 - A_s/R_C - 2(R_B/R_C)^4/3]^2$ and Γ_{atomic}, defined by Eq. (1), is the rate for the recoil heating for the atomic case. For red detuning

$$\Gamma^{\text{red}}_{\text{binary}} = \Gamma^{(1)}_{\text{binary}} \sum_v \frac{\nu_v\gamma_v}{(\Delta_A - \Delta_v)^2 + (\gamma_v/2)^2}\,. \tag{12}$$

Figure 2 shows the ratio η for $x = 1$, defined as

$$\eta^{\text{color}} = \frac{\Gamma^{\text{color}}_{\text{binary}}/n\lambda\!\!\!^{-3}_A}{\Gamma_{\text{atomic}}}\,, \tag{13}$$

where color stands for red or blue, depending on which detuning case we consider. This factor shows how the binary loss and pure atomic loss compare when we have one particle in a volume $\lambda\!\!\!^{-3}_A$. This density scale can be regarded as more or less typical for condensates. We should emphasize that the pure ratio $\Gamma^{\text{color}}_{\text{binary}}/\Gamma_{\text{atomic}}$ is proportional to the particle density in the condensate, as expected for binary processes. Figure 2 shows clearly the differences between the blue and red detuning cases. The detuning region displayed is one where the reflection principle approach can be applied, and where the condition $|\Delta_A| \gg \gamma_{\text{con}}$ is fulfilled. We have used a model Na_2 system, with $A_s = 84.3a_0$, $\gamma_v = 20.0$ MHz, $R_B = 42.0a_0$, and $C_3 = \pm 10.0$ atomic units. We have also set $b_C = 2/\sqrt{3}$ and $f_3 = 1/2$, yielding

$$\eta^{\text{blue}} = \left(\frac{16\pi^2}{9}\right) g_C\,, \tag{14}$$

$$\eta^{\text{red}} = \eta^{\text{blue}} \sum_v \frac{\nu_v\gamma_v}{(\Delta_A - \Delta_v)^2 + (\gamma_v/2)^2}\,. \tag{15}$$

Our results show several important features which will show up in optical probing of a condensate. First, the binary rate at typical condensate densities can easily exceed

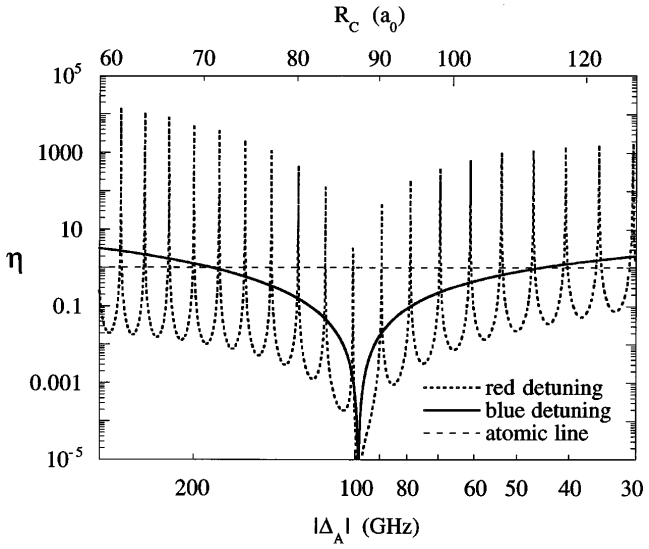

FIG. 2. The ratio of the light-induced loss rates for atoms in the condensate as a function of the detuning (here $x = 1$). The horizontal dashed line stands for the case when $\Gamma_{\text{binary}} = \Gamma_{\text{atomic}}(n\lambda_A^3)$, i.e., $\eta = 1$. The red-detuned case shows strong modulation of the loss rate due to the bound state resonances, but both the red- and blue-detuning cases feature clearly the suppression of the binary loss when the Condon point hits the node of the ground state wave function at $|\Delta_A| \sim 100$ GHz.

the atomic rate, depending on detuning. The rate for blue detuning varies smoothly with frequency and is comparable to the atomic one. Second, the rate for red detuning exhibits sharp features, just like the well-known photoassociation spectra of cold alkali gases [11–16], and can exceed the atomic rate by orders of magnitude. These spectra can be expected to provide a sensitive probe of atomic interactions in the condensate. They are very sensitive to the ground state wave function of the pair, that is, the pair correlation function, and can be used to locate the position of the node in Ψ_g near $R \approx A_s$ (for positive A_s). Spectra in a condensate will be simpler than for the $\sim 500\ \mu$K traps where photoassociation spectra have heretofore been measured, since only s waves contribute and there is no thermal broadening. Thus they provide the opportunity for a more precise determination of A_s, and for probing possible modifications in the pair correlation function due to the presence of the condensate. In addition, the detailed line shapes of the spectra [17] will likely be broadened and shifted by three-body or higher order interactions. Therefore, a careful study of line shapes versus condensate fraction x is likely to provide a much needed experimental tool for investigating such interactions. Real alkali molecules also have several excited molecular states that have photoassociation spectra, so a wide range of Condon points can be surveyed, and provide consistency checks for probing Ψ_g. The hyperfine structure in such spectra can now be accurately calculated [21], as well as hyperfine radiative line strengths [22], and the expression in Eq. (11) is readily generalized to include the hyperfine structure.

In this Letter we have shown that, for reasonable detunings from the atomic resonance, excitation in binary collisions will produce marked and characteristic modifications to the lifetime of the condensate. The frequency dependence of the photodestruction rate of the condensate depends sensitively on the ground state wave function. We have given simple analytic formulas for estimating the condensate spectrum in the extreme quantum limit of very low temperature evaporatively cooled gases.

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[1] M. H. Anderson *et al.,* Science **269**, 198 (1995).
[2] C. C. Bradley *et al.,* Phys. Rev. Lett. **75**, 1687 (1995).
[3] K. B. Davis *et al.,* Phys. Rev. Lett. **75**, 3969 (1995).
[4] C. S. Adams *et al.,* Phys. Rep. **240**, 143 (1994).
[5] Y. Castin *et al.,* in *Bose-Einstein Condensation,* edited by A. Griffin *et al.* (Cambridge Univ. Press, Cambridge, 1995), p. 173, and references therein.
[6] H. M. Wiseman and M. J. Collett, Phys. Lett. A **202**, 246 (1995).
[7] R. J. C. Spreeuw *et al.,* Europhys. Lett. **32**, 469 (1995).
[8] L. You *et al.,* Phys. Rev. A **53**, 329 (1996), and references therein.
[9] J. Javanainen, Phys. Rev. Lett. **72**, 2375 (1994); **75**, 1928 (1995); J. Javanainen and J. Ruostekoski, Phys. Rev. A **52**, 3033 (1995); R. Graham and D. Walls, Phys. Rev. Lett. **76**, 1774 (1996).
[10] O. Morice *et al.,* Phys. Rev. A **51**, 3896 (1995).
[11] L. P. Ratliff *et al.,* J. Chem. Phys. **101**, 2638 (1994).
[12] R. A. Cline *et al.,* Phys. Rev. Lett. **73**, 633 (1994).
[13] P. D. Lett *et al.,* Ann. Rev. Phys. Chem. **46**, 423 (1995).
[14] R. Cote *et al.,* Phys. Rev. Lett. **74**, 3581 (1995).
[15] J. R. Gardner *et al.,* Phys. Rev. Lett. **74**, 3764 (1995).
[16] E. R. I. Abraham *et al.,* J. Chem. Phys. **103**, 7773 (1995).
[17] R. Napolitano *et al.,* Phys. Rev. Lett. **73**, 1352 (1994).
[18] W. E. Parry, *The Many-Body Problem* (Oxford Univ. Press, Oxford, 1973), Sec. 4.3.
[19] H. T. C. Stoof, Phys. Rev. A **49**, 3824 (1994).
[20] P. S. Julienne, J. Res. Nat. Inst. Standards Tech. (to be published).
[21] C. J. Williams *et al.,* Phys. Rev. A **53**, R1939 (1996).
[22] E. Tiesinga *et al.,* J. Res. Nat. Inst. Standards Tech. (to be published).
[23] H. T. C. Stoof *et al.,* Phys. Rev. A **39**, 3157 (1989).
[24] F. H. Mies and P. S. Julienne, IEEE J. Quant. Electron. **15**, 272 (1979).
[25] Yu. Kagan *et al.,* JETP Lett. **48**, 56 (1988) [Pis'ma Zh. Eksp. Teor. Fiz. **48**, 54 (1988)].