Quantal Treatment of Cold Collisions in a Laser Field

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(Received 24 June 1993)

We study the kinetic energy changes taking place in collisions of atoms in a laser cooled cloud. We present a quantum approach, which can treat cold collisions at temperatures well below the Doppler limit of laser cooling. In our numerical treatment we observe an increase in the kinetic energy which depends strongly on the laser detuning and intensity.

PACS numbers: 32.80.Pj, 32.80.Bx, 34.50.Rk

The densities now achievable by laser cooling mean we have to look into the effect of interactions between ultracold atoms. This is especially true as the collisions occur in the presence of laser fields, i.e., the fields that are doing the cooling and trapping. We need in particular to determine how these collisions limit the densities and temperatures that one can achieve. Proposed methods of achieving Bose-Einstein condensation based on alkali atoms are critically dependent on the limiting densities obtainable in laser cooling schemes [1].

There have been several studies of how long range collisions inhibit laser cooling and produce excess heating via extra diffusion [2]. These analyses use semiclassical methods and in their most efficient form simply determine the diffusion coefficient for an atom with a static partner using the quantum regression theorem. They do not treat the quantal motion of the atoms and are not able to treat correctly the important region of shorter ranges, where the dipole-dipole force tunes the atomic binary system into resonance with the field. This is particularly important for the production of kinetic energy in the relative motion [3–7]. The only quantal treatment of this problem that we are aware of does not treat the effect of population recycling [8]. Fortunately, we have now been able to exploit the recently developed Monte Carlo method to produce a fully quantum treatment of this critical issue.

There are two possible mechanisms for excitation in long range collisions to produce heating. First, a pair of atoms may undergo a fine structure changing collision while in the excited state. This leads to an increase in the kinetic energy of the pair. Second, spontaneous decay can take the excited system back to the ground state, retaining the kinetic energy increase gained by rolling down the excited state potential. In this Letter we study the latter effect, i.e., radiative heating. We focus on the bulk of collisions that produce moderate heating, rather than on the less likely but still important collisions that produce major kinetic energy changes and trap loss.

We consider the translational wave packets of the atomic system on the ground and excited states coupled by the laser field. We solve numerically the coupled time-dependent two-state Schrödinger equation. The spontaneous decay is included by using the stochastic Monte Carlo method [9,10], which allows random quantum jumps from the excited state to the ground state. Individual time-dependent wave functions can then be combined and in the limit of a large ensemble should agree with treatment based on the density matrix. This method gives an enormous advantage in comparison with a direct method based on the density matrix and enables us to do a calculation of the properties of an open system which otherwise would not be computable given the core memory of present machines.

We decompose the three dimensional combined wave function for the two colliding atoms into partial waves corresponding to different angular momenta and consider a simple model of two levels. We ignore hyperfine structure, the spatial dependence of the dipole moment, and the precise structure of the inner core, including deviations of the excited state structure from the dipole-dipole potential $-C_3/R^3$, where R denotes the interatomic distance. However, the effect of the centrifugal barrier $\hbar^2 l(l+1)/2\mu R^2$ on both the ground state and excited state is included in order to estimate the cross section of the collisions leading to heating, although $l \to l' \neq l$ transitions are still ignored. We consider a temperature region which is well below the Doppler limit of laser cooling, but far enough above the recoil limit so that we can ignore photon recoil effects on the dynamics.

We apply our model to the ${}^2S_{1/2} \to {}^2P_{3/2}$ transition in a cesium quasimolecule, between the fine structure states 0_g^+ (ground) and 0_u^+ (excited). An example of the level configuration for certain values of the laser field detuning Δ (atomic resonance frequency minus laser frequency), coupling strength Ω, and angular momentum quantum number l is given in Fig. 1. The ground state level has been raised by the frequency of the laser field so that the resonance point becomes a bare state level crossing. We scale our spatial variables with the transition wavelength λ over 2π: $\lambda \simeq 136$ nm; this distance unit is equal to $2560a_0$, where a_0 is the Bohr radius. Our energy unit is $\hbar^2 k_\lambda^2/2\mu = 2.74 \times 10^{-30}$ J; μ is the reduced mass of the system and $k_\lambda = 1/\lambda$.

For the dipole moment we take the molecular values of Ref. [5]. In our scaling the atomic linewidth is $\Gamma_{\rm at} = 1240$ and the dipole force factor is $C_3 = 1.55\Gamma_{\rm at}$. The molecular linewidth is $\Gamma = 4\Gamma_{\rm at}/3$. Altogether the scaled

0031-9007/94/72(15)/2367(4)$06.00

Schrödinger equation to be solved is, in the absence of fluorescence,

$$i\frac{\partial}{\partial t}\begin{pmatrix}\Psi_g\\ \Psi_e\end{pmatrix}=\begin{pmatrix}-\frac{\partial^2}{\partial R^2}-\Delta+\frac{l(l+1)}{R^2} & \Omega\\ \Omega & -\frac{\partial^2}{\partial R^2}-\frac{C_3}{R^3}+\frac{l(l+1)}{R^2}\end{pmatrix}\begin{pmatrix}\Psi_g\\ \Psi_e\end{pmatrix}, \tag{1}$$

where Ψ_g and Ψ_e are the probability amplitudes for the bare ground (g) and bare excited state (e). The kinetic energy operator $w_{\rm kin}$ in our scaling is the square of the relative momentum operator p, which scales with $\hbar k_\lambda$.

Our study consists of simply propagating a Gaussian wave packet, initially in the bare ground state, into the crossing region and the inner core area behind it, and eventually back to the area far from both the inner core and the crossing region. Although in many cases the field-dressed, i.e., adiabatic, states form a more natural basis to work with than the bare states, we prefer the latter. The inclusion of quantum jumps, needed in the Monte Carlo simulations of dissipation, is simpler in this basis. The crossing region is usually taken to be the area where the adiabatic states and the bare states differ strongly (see Fig. 1). In the momentum space our initial wave packet has a mean $p_0 = -10$ and a width $\Delta p_0 = 2$. Eventually one needs to determine the collisional momentum change matrix $W(p_0, p)$ for different p_0 and vanishing Δp_0, but for the moment we only look at a single p_0 and concentrate on the effect of other parameters on heating.

Because of the crossing and random quantum jumps, there are several "paths" the wave packet can take as it enters the interaction region surrounding the crossing point, becomes later reflected by the inner core, and eventually traverses the crossing region again on its way out. Typically most of the packet passes smoothly from the ground state to the excited state as it traverses the crossing region, and becomes rapidly accelerated under the influence of the $-C_3/R^3$ potential. However, before it gets too far, a quantum jump brings it back to the ground state. After a reflection at the core, the packet reenters the crossing region. If it has been sufficiently accelerated, it experiences substantially less excitation than during the first passage. It is important to note that phase effects, such as Stückelberg oscillations, are absent due to the spontaneous decay.

The results obtained by using the Monte Carlo method are only approximations of the density matrix result. The correspondence improves as the number of members (N) in the ensemble is increased. Our experience is that the characteristic behavior is obtained with about $N \sim 50$, although peaks of individual wave packets are still slightly visible in the final result. However, to smooth out the spikes requires ensemble sizes around $N \gtrsim 500$; such sizes are not practical when one intends to span a large and multidimensional parameter space. A discussion of suitable ensemble sizes and a review of both the master equation and the Monte Carlo methods for wave packet dynamics can be found in Ref. [10].

If we were working at Doppler temperatures, where the mean kinetic energy of the colliding pair is much larger, we could apply the Landau-Zener theory which neglects spontaneous decay in the crossing region of the potential surfaces. At the recoil temperatures we consider in this Letter, spontaneous decay cannot be ignored in the curve crossing regions, and the simple Landau-Zener model gives poor quantitative predictions. It does, however, provide insight into the parameter dependence of the heating mechanism on the detuning Δ and coupling strength Ω. In this approximation the probability to be excited as the crossing region is traversed is [11–13]

$$P_{\rm LZ} = 1 - \exp\left(-\frac{\pi\Omega^2}{|p|\alpha}\right) \tag{2}$$

for each momentum component p in the ground state wave packet. Here $\alpha = 3(\Delta^4/C_3)^{1/3}$ is the absolute slope of the excited state potential at the crossing point (all parameters are given in scaled units). The excited components then accelerate down the slope towards the core and in a dissipative system are eventually brought back to the ground state by spontaneous decay.

In Fig. 2(a) and Table I we present examples of our results which show that clear increases in the mean and the width of the momentum distribution occur after a

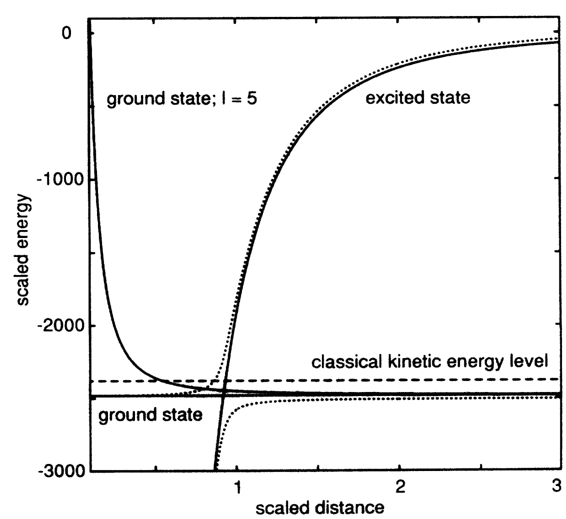

FIG. 1. Here we show an example of the level configurations studied in this Letter. The solid lines correspond to the bare states and the dotted lines to the field dressed, i.e., adiabatic states. The centrifugal barrier on the ground state for $l = 5$ is also given. The dashed horizontal line shows the level of kinetic energy corresponding to the classical equivalent of our initial wave packet with $p_0 = -10$. The specific parameters used here are $\Delta = 2\Gamma_{\rm at}$ and $\Omega = \Gamma_{\rm at}/5$.

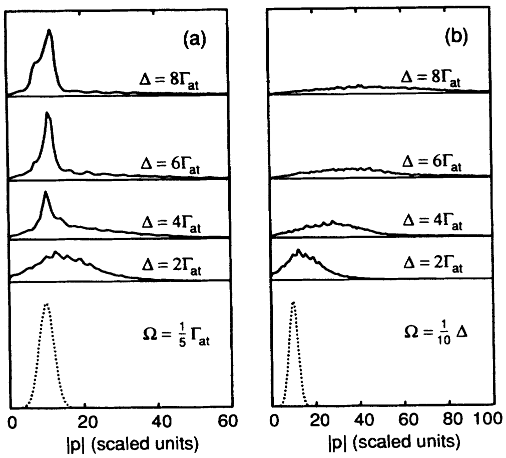

FIG. 2. Here we present the final distributions of absolute momentum $|p|$ (solid lines), for different values of detuning Δ and coupling strength Ω. In (a) we present the case where Δ is changed but Ω is kept constant. In (b) they are both changed, but the ratio Ω/Δ is always equal to 0.1. The dotted line at the bottom of both (a) and (b) is the initial distribution. In (a) the ensemble sizes N are 121, 136, 65, and 52, in order of increasing Δ, and similarly in (b) 121, 50, 53, and 62.

cold collision, even for a modest coupling strength Ω. As the detuning is increased, the heating saturates, because the increase in the slope (and hence acceleration) in the excited level is counteracted by the diminished excitation probability, as indicated by Eq. (2). The number of quantum jumps per ensemble member decreases radically as detuning is increased, since the overall size of the excited population determines the probability for a quantum jump to take place. As the portion of no-jump cases becomes appreciable the final momentum distribution becomes a combination of a large, unaffected component and a small but wide tail.

If we increase both the detuning and the coupling together so that the ratio between them is maintained, we see a substantial increase in kinetic energy change [Fig. 2(b) and Table II]. As the detuning is increased, the crossing point moves closer to the core and the increase in the slope of the excited state potential has a strong effect on the acceleration. However, since the coupling is also increased, the excitation probability does not fall very rapidly with the increase in detuning. Hence a major part of the wave packet always experiences strong acceleration.

In our approximation the repulsive ground state core is represented by a solid wall at $R = 0$ when $l = 0$. The nonzero values of l affect the excited state very little, but on the ground state a clear barrier with a classical turning point $R_l \simeq \sqrt{l(l+1)}/|p_0|$ appears. The effect of different values of l on heating with certain combinations of detuning and coupling is shown in Fig. 3. When l is increased, this point moves outward and eventually meets the crossing point. As expected, we observe then a

TABLE I. Here we show the additional data for the case where Ω has the constant value $0.2\Gamma_{at}$, but Δ changes. The ensemble size is N; the number n of quantum jumps in each simulation run within the ensemble has the average $\bar{n}$ and extreme values n_{min} and n_{max}. The percentage of no-jump members in the ensemble is given in the seventh column. The change $\Delta\langle w_{kin}\rangle$ in the semiclassical kinetic energy is normalized to the initial kinetic energy; also, it is calculated and normalized using the distributions of Fig. 2(b) only up to $|p| = 100$.

Δ/Γ_{at}	l	N	$\bar{n}$	n_{min}	n_{max}	$n = 0$ (%)	$\Delta\langle w_{kin}\rangle$
2	5	51	12	4	19	0	3
4	5	52	4	1	13	0	6
6	0	65	2	0	10	18	6
8	0	52	1.2	0	4	31	6

rapid reduction in the heating, and a rough cutoff value l_{max} can be obtained. For values of l larger than l_{max} the barrier simply reflects the incoming packet before the crossing region is reached.

Collisions introduce an extra diffusion term which can be estimated by $D_c = 2\pi n|p_0|\sum_{l=0}^{l_{max}}(2l+1)\Delta\langle w_{kin}\rangle$, where n is the density of atoms per cubic wavelength. This can be compared with the diffusion coefficient $D_s = 3\Omega^2/4\Gamma_{at}$ for Sisyphus cooling at large detuning [14]. For the detuning $\Delta = 8\Gamma_{at}$ and driving field $\Omega = 0.8\Gamma_{at}$, the simulation results suggest values of $l_{max} = 10$ and $\Delta\langle w_{kin}\rangle = 25$. The density at which the two diffusion coefficients are equal is then 2×10^{12} cm^{-3}. This is only a rough estimate of the density at which collisional heating will become important since the collisions produce a non-Maxwellian distribution of atomic velocities. More simulations for different values of p_0 are required before one can fully map our results into the thermodynamics of the trapped and continuously cooled atom cloud. Many of the details mentioned earlier and ignored in our treatment can be added in order to make it correspond better with specific systems, although then some generality will be lost. Also, the effect of the centrifugal barrier may vary strongly for different transitions and types of atoms. By allowing a third level one might be able to study both the fine structure heating and radiative heating within a single quantum model. Then one needs detailed information of the potentials all the way between the core and the crossing region. Wave packet dynamics in the inner core region has very different numerical requirements than at

TABLE II. The additional data corresponding to Fig. 2(b). Notation is the same as in Table I.

Δ/Γ_{at}	Ω/Γ_{at}	N	$\bar{n}$	n_{min}	n_{max}	$\Delta\langle w_{kin}\rangle$
2	0.2	51	11	3	19	3
4	0.4	50	12	4	23	12
6	0.6	53	12	6	25	20
8	0.8	62	14	5	33	27

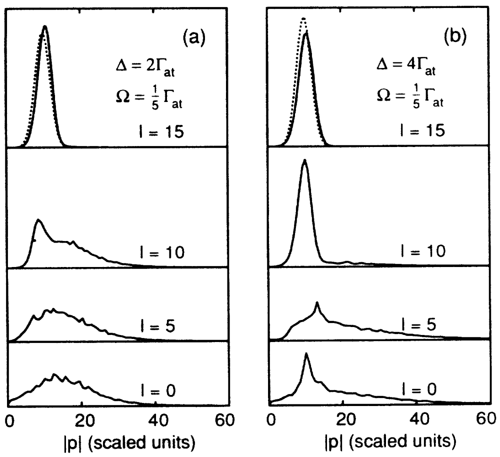

FIG. 3. Here we present the final distributions of absolute momentum $|p|$ for some values of l, and for two detunings. The dotted line, presenting the initial distribution, is drawn overlapping with $l = 15$ values, in order to show the small but finite changes. In (a) the ensemble sizes are 121, 51, 54, and 13, in order of increasing l. Similarly in (b) we have $N = 136$, 52, 50, and 15. The crossing point is given by the expression $R_{cr} = (C_3/\Delta)^{1/3}$, and has values $R_{cr} = 0.92$ (a) and $R_{cr} = 0.73$ (b). The classical turning points for different values of l are, for both (a) and (b), $R_5 = 0.55$, $R_{10} = 1.05$, and $R_{15} = 1.55$. The value for l_{max} is expected to decrease as the detuning Δ is increased; such behavior becomes clearly visible when (a) and (b) are compared.

the long range crossing region, so their unification under a single scheme is far from trivial. We have demonstrated a novel theoretical technique that shows how collisions in a laser field limit the densities one can reach at particular temperatures. This has important consequences for a whole range of experiments (e.g., Bose-Einstein condensation) planned with laser cooled atoms.

This research was supported by the U.K. Science and Engineering Research Council. M.H. acknowledges support from the Royal Society. K.-A.S. is grateful to the Academy of Finland and Suomalainen Tiedeakatemia for financial support. We would like to thank John Cooper, Chris Foot, and Paul Julienne for constructive and helpful discussions.

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