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LETTER TO THE EDITOR

Counting cold collisions

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Abstract. We have explored experimentally a novel possibility to study exoergic cold atomic collisions. Trapping of small countable atom numbers in a shallow magneto-optical trap and monitoring of their temporal dynamics allows us to directly observe isolated two-body atomic collisions and provides detailed information on loss statistics. A substantial fraction of such cold collisional events has been found to result in the loss of one atom only. We have also observed for the first time a strong optical suppression of ground-state hyperfine-changing collisions in the trap by its repump laser field.

The observation of physical phenomena at the atomic level often provides new insights into the details of the processes under study, usually hidden in ensemble samples. Trapping of individual neutral atoms in a magneto-optical trap (MOT) [1] realized in [2, 3] allowed us to obtain information on the internal and external atomic dynamics in the trap with excellent contrast [4]. Cold inelastic collisions are usually associated with high atomic densities and consequently with experiments on large numbers of trapped atoms. Here we study them with only a few atoms. In this situation one is able to monitor the instantaneous number of trapped atoms exactly and observe isolated load and loss events. In figure 1 we show an example of the dynamics of the trapped atom number in the MOT operating at constant conditions. Such a 'digitized' signal provides detailed information on the collisional statistics we will analyse in this letter.

By far the most popular choice for collision studies is the MOT providing dense samples of cold atoms. An exoergic collision converts internal atomic energy to kinetic energy equally divided between the colliding partners. If the transferred kinetic energy is greater than the recapture ability of the trap, the collision leads to trap loss. Here we experiment with a very shallow trap which is sensitive to low-energy collisional processes. It enables us to observe a strong optical suppression of ground-state hyperfine-changing collisions by the MOT repump laser field and to infer the corresponding rate constant.

There has been extensive progress in both experimental and theoretical investigations of collisions between laser-cooled atoms [5]. The main method of observing collisions used so far has been to abruptly change experimental parameters (usually switching off or on an atomic beam loading the trap) and to watch the trap population decay or increase. The rate equation for the total number, N, of trapped atoms is given by $\dot{N} = R - N/\tau_{\mathrm{coll}} - \beta \int n^2(\boldsymbol{r}, t)\, \mathrm{d}^3 r$, where the first two terms on the right-hand side describe the loading rate R and the loss rate due to collisions with background molecules, respectively. The last term represents the intra-trap collisional loss rate due to collisions between trapped atoms with a density profile $n(\boldsymbol{r}, t)$. In our experiments the number of trapped atoms is small and can be determined exactly.

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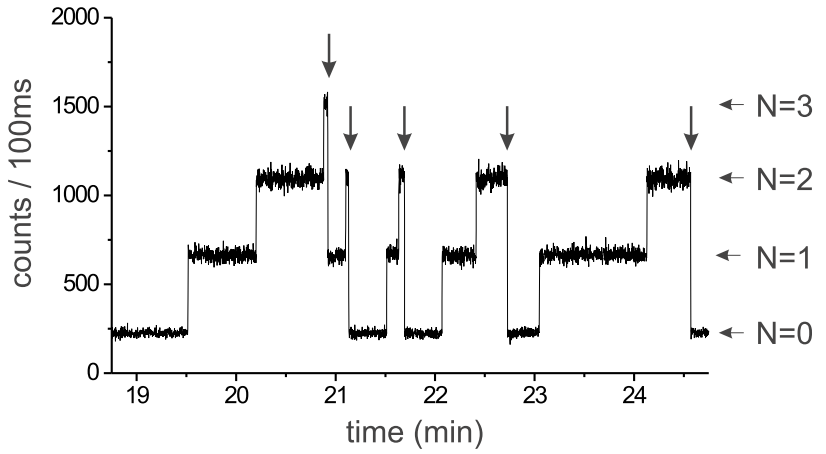

Figure 1. Excerpt from a typical MOT fluorescence signal observed with an avalanche photodiode. Five isolated cold collisions (two-atom losses) (the first direct observation of isolated two-body atomic collisions in a MOT was reported in [7]) are shown (arrows). The fluorescence level at atom number $N = 0$ corresponds to residual stray light. Clearly separated fluorescence steps can be easily resolved for atom number up to $N = 20$.

Thus information can be obtained from temporally resolved fluctuations in the atom number under conditions of dynamical equilibrium providing a possibility to work under constant experimental parameters.

The number of trapped atoms and its temporal fluctuations is determined by the balance between loading the MOT from the low-pressure atomic vapour and different loss mechanisms, removing one or two atoms from the trap with the corresponding loss event rate $L_{1\text{atom}}(N)$ and $L_{2\text{atoms}}(N)$ at atom number N, respectively,

$$\dot{N} = R - L_{1\text{atom}}(N) - 2L_{2\text{atoms}}(N). \tag{1}$$

We can distinguish various events with 100% contrast and thus all terms on the right-hand side can be now measured independently. The two-atom losses obviously arise from collisions of trapped atoms $2L_{2\text{atoms}}(N) = \beta N(N-1)/V$, where $V = (\pi/2)^{3/2} r_0^3$ is the effective trapping volume. A simple guess is also to assume that $L_{1\text{atom}}$ in (1) is associated with collisions with background molecules $L_{1\text{atom}}(N) = N/\tau_{\text{coll}}$. In order to check this assumption we have plotted in figure 2 the N dependence of R, $L_{1\text{atom}}$ and $L_{2\text{atoms}}$ by counting the corresponding load and loss events for each atom number under constant conditions. As expected we have found R to be independent of N and the two-atom loss rate to scale quadratically with the atom number. However, and surprisingly, a substantial part of the one-atom loss rate also scales quadratically with N indicating 'soft' two-body collisions resulting in the loss of only one atom†. So we can rewrite (1) with $L_{1\text{atom}}(N) = N/\tau_{\text{coll}} + \beta_{1\text{atom}} N(N-1)/V$ and $2L_{2\text{atoms}}(N) = \beta_{2\text{atoms}} N(N-1)/V$ in the form

$$\dot{N} = R - N/\tau_{\text{coll}} - \beta N(N-1)/V \tag{2}$$

† We also checked that R and $1/\tau_{\text{coll}}$ increase with increasing Cs background pressure, while $\beta_{1\text{atom}}$ and $\beta_{2\text{atoms}}$ remain constant.

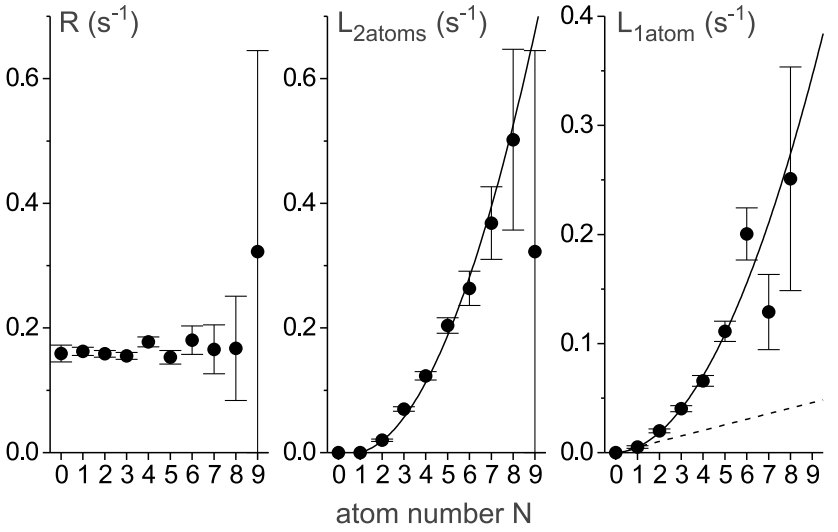

Figure 2. Load and loss event rates as a function of the atom number N measured for a total cooling laser intensity of $I = 42$ mW cm^{-2} and detuning $\delta = -3.35\Gamma$ and average atom number during the measurement $\langle N \rangle = 2.6$. Full curves represent a second-order polynomial fit. The broken curve shows the linear dependence due to collisions with the background gas. Error bars indicate the statistical error of load and loss occurrences.

with $\beta = \beta_{2\text{atoms}} + \beta_{1\text{atom}}$. Both loss coefficients can be determined with good accuracy by quadratic fitting of the corresponding N dependences.

Our experimental set-up has been described in detail elsewhere [3, 4]. A six-laser-beam σ^+–σ^- MOT is loaded from a low-pressure caesium vapour (at a base pressure of better than 10^{-10} mbar). The trap laser detuning δ from the Cs cooling transition $F = 4 \rightarrow F' = 5$ (typically $\delta = -3\Gamma$, in terms of the natural linewidth $\Gamma = 2\pi \times 5.2$ MHz) is precisely controlled by a heterodyne phase-locking technique. To prevent the atom escaping from the cooling cycle by a decay into the $F = 3$ ground state, a second (repump) laser is introduced, resonant with the $F = 3 \rightarrow F' = 4$ transition and stabilized by standard techniques to $\Gamma/4$. The MOT magnetic quadrupole field is produced by permanent magnet discs with tunable field gradients up to $B' = \mathrm{d}B_z/\mathrm{d}z = 800$ G cm^{-1}. Stiff magnetic field gradients lead to a strong reduction of the capture rate [6]. It was also found that the MOT spring constant is linearly dependent on the quadrupole magnetic field gradient, while the temperature of trapped atoms is comparable to a low-field MOT [6, 7]. Thus the atomic density is expected to be proportional to $(B')^{3/2}$ and as a result the probability for collisions between trapped atoms rises dramatically $\propto (B')^3$. For $B' = 375$ G cm^{-1} in our experiment the two-atom loss probability is comparable with the probability for background collisions: a comfortable situation for a measurement. The spatial distribution of the MOT fluorescence measured by a CCD camera has a Gaussian distribution with $1/\mathrm{e}^2$ radius r_0 between 7 and 24 μm depending on the laser intensity. The trap size was observed to be independent of the atom number (up to $N = 8$) ensuring that radiation trapping effects [8] can be ignored. Fluorescence of the atoms trapped in the MOT is observed with avalanche photodiodes in single-photon counting mode. Typical photon counting rates are 3–20 kHz/atom depending on the trap laser detuning and intensity. The probability for eventual misinterpretations (for example, two one-atom losses occurring simultaneously within our integration time

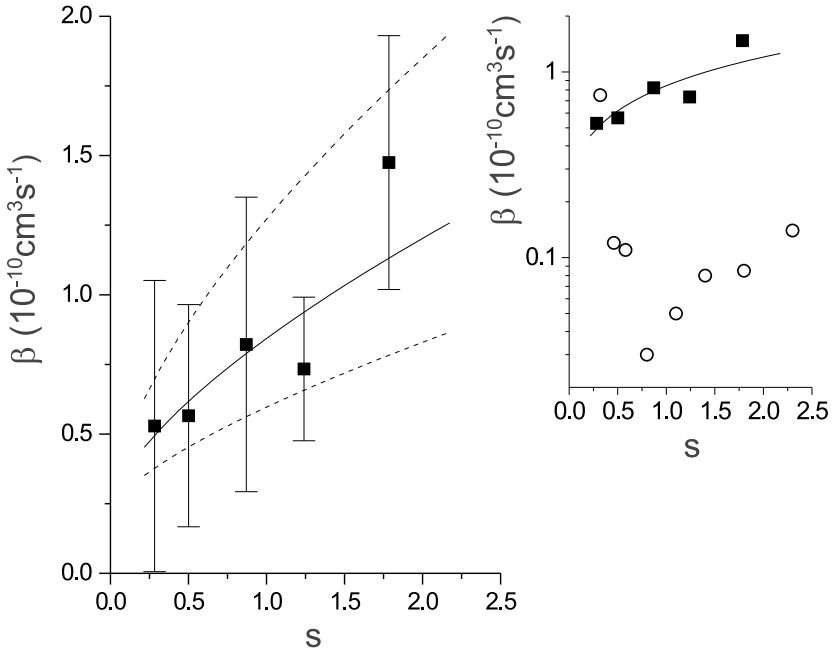

Figure 3. Total collisional loss coefficient β versus the cooling laser saturation parameter s in our trap (squares). Full curve, fit according to the semiclassical model (see text); dotted curves, uncertainty in the trap depth of a factor of two. Inset, same (log) plot in comparison with standard Cs-MOT data (circles show an average over the scattered data in [9]).

of 100 ms and detected as a two-atom loss event) is below 1% and can be neglected here.

Three main exoergic collisional processes in MOTs have been identified [5]: the fine-structure-changing collision (FCC) is represented by $\mathrm{A} + \mathrm{A} + \hbar\omega \rightarrow \mathrm{A}_2^*(\mathrm{P}_{3/2}) \rightarrow \mathrm{A}^*(\mathrm{P}_{1/2}) + \mathrm{A} + \Delta E_{\mathrm{FCC}}$ with the energy $\Delta E_{\mathrm{FCC}}/2$ transferred to each atom. For Cs atoms $\Delta E_{\mathrm{FCC}}/2k_{\mathrm{B}} \approx 400$ K and FCC collisions ultimately cause an escape of both atoms from the MOT, usually no more than 1 K deep. For radiative escape (RE), spontaneous emission of a photon redshifted from the atomic resonance takes place during the collision. The process is described by $\mathrm{A} + \mathrm{A} + \hbar\omega \rightarrow \mathrm{A}_2^* \rightarrow \mathrm{A} + \mathrm{A} + \hbar\omega'$ with an energy of $\hbar(\omega - \omega')/2$ transferred to each atom. The resulting kinetic energy is continuously distributed and the corresponding loss rate is sensitive to the effective trap depth. Exoergic hyperfine-changing collisions (HCCs) on the molecular ground state can also lead to losses if the trap is sufficiently shallow. For Cs, a change from $6\mathrm{s}\ ^2\mathrm{S}_{1/2}(F = 4)$ to $6\mathrm{s}\ ^2\mathrm{S}_{1/2}(F = 3)$ in one of the colliding atoms transfers about $\Delta E_{\mathrm{HCC}}/2k_{\mathrm{B}} = 0.22$ K to each atom. For interpretation of trap-loss measurements it is essential to distinguish which collision processes are producing the trap loss. One such possibility is to use the intensity dependence of different loss channels. In a standard Cs MOT Sesko *et al* [9] observed a rapid increase in β as the intensity I of the trapping laser dropped below $s = 0.8$, see figure 3 (data scaled to the effective saturation parameter $s = I/I_{\mathrm{S}}/[1 + (2\delta/\Gamma)^2]$). They interpreted this loss as being due to hyperfine-changing collisions, but could not infer β_{HCC} directly because even at the lowest MOT laser

intensity the trap loss does not become independent of the trap depth. The rate constant β_{HCC} has been estimated at between 10^{-10} and 10^{-11} cm^3 s^{-1}. For higher intensities the trap becomes too deep for atoms to escape after HCC and this contribution to the trap losses disappears. On the other hand, the probability for one of the colliding atoms to be excited to an attractive potential curve (corresponding to the $S_{1/2} + P_{3/2}$ asymptote) increases with growing laser intensity and FCC and RE collisions constitute the main loss mechanism for usual traps. This interplay of different loss channels imparts a characteristic form to the intensity dependence of β also observed in MOTs operating with other alkalis [5]. In our trap we observe a similar loss rate coefficient at low excitation rates but the measured dependence in s is quite different (figure 3). In particular, the steep rise in β at low s is absent.

It can be easily explained by a substantially reduced 'trap depth' ΔU (or more accurately by the maximum initial kinetic energy enabling an escape) due to large magnetic field gradients B' in our MOT. The effective 'deceleration distance' (where the scattering light force acting on the escaping atom is close to its maximum resonant value) scales as $1/B'$. Numerical simulations [10] substantiate the interpretation that ΔU in figure 3 is a weak function of the trap laser intensity [11] and is always smaller than the energy gained in a HCC. As a result every ground-state HCC process leads to a two-atom loss and the HCC contribution (about 50% for low intensity in figure 3) to the total trap losses is nearly independent of the trap laser intensity. In a shallow trap the RE contribution to the total trap losses is also substantially larger in comparison to usual MOTs and is dominant at higher laser intensities. Based on [12] we have performed semiclassical numerical calculations of the distribution of the energy gained as a result of an RE collision for the parameters of our experiment giving reasonable agreement with our measurements (see figure 3). This allows us to infer the value for the HCC collisional loss coefficient, 2.0×10^{-11} cm^3 s^{-1}, as an intensity-independent offset in β. However, as we will see below, ground-state collisions can be strongly affected by the repump laser.

As already mentioned a significant part (typically 10% of the total loss rate) of two-body collisions in our trap lead to one-atom losses. The energy gained in a collision is equally divided between both atoms (the initial kinetic energy of colliding atoms of the order of some 100 μK can surely be neglected). For producing a one-atom loss this energy must be comparable to the effective trap depth. Thus one-atom losses obviously come from RE processes. To be recaptured, an escaping atom with an initial kinetic energy of 0.1 K has to scatter about 10^3 photons and thus the statistical fluctuation of the trap depth is about 3%. Statistical fluctuations also lead to deviations from originally counterpropagating straight-line paths of atoms leaving the trap which may be important because of the anisotropy of the trapping potential. As shown numerically [11] and experimentally [13], ΔU varies by as much as a factor of 4 between the shallowest and deepest directions. It can be estimated [10] that for RE collisions releasing more energy than 0.1 K about 50% of collided atom pairs gain kinetic energy in the interval between 0.1 and 0.4 K. However, even under these assumptions the frequent occurrence of one-atom losses observed in the experiment cannot be explained and needs some additional analysis [10].

The 9 GHz blue detuning of the MOT repump laser is so large that it cannot exert a force on a single escaping atom and thus cannot effect the trap depth. However, we have observed a strong dependence of the loss rates on the repump laser intensity, figure 4. We explain the exponential decay of the loss rates to a nearly constant value at high intensities by the process of optical shielding first reported in [14] and further investigated in [15].

At small interatomic distance $R_{\mathrm{C}} \approx 100$ Å the repump laser is resonant with the repulsive quasimolecular potential corresponding to the asymptotic state $|S + P\rangle$. In the dressed-atom

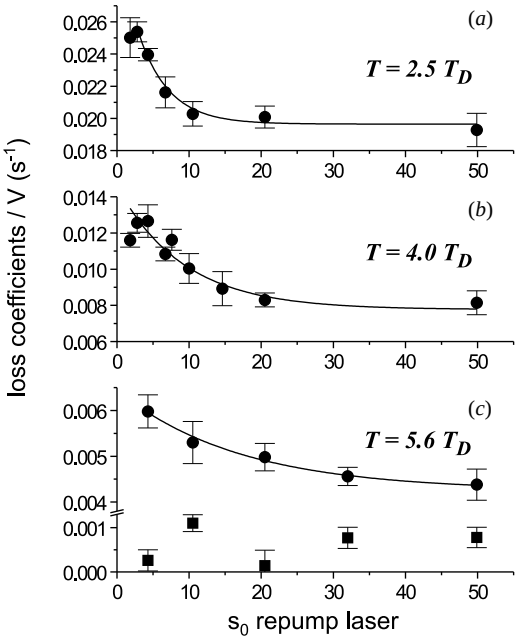

Figure 4. Loss rates (circles, $\beta_{2\text{atoms}}/V$; squares, $\beta_{1\text{atom}}/V$) as a function of the repump laser intensity for different cooling laser parameters: (*a*) $s = 0.87$, (*b*) $s = 1.74$ and (*c*) $s = 5.19$ with representative values for $\beta_{1\text{atom}}/V$. Full curves are calculated from the Landau–Zener model (see text).

picture [14, 15] the presence of the resonant laser field leads to an avoided crossing between the states $|\mathrm{S} + \mathrm{S}, n_\gamma\rangle$ and $|\mathrm{S} + \mathrm{P}, n_\gamma - 1\rangle$ with a Rabi splitting $\hbar\Omega$ at the Condon point R_C. Here n_γ is the photon number in the repump laser field. This splitting prevents the atom pairs from approaching in the $|\mathrm{S} + \mathrm{S}\rangle$ state close enough for the ground-state collisions to occur. In our measurements the repump laser intensity is varied from $s_0 = 2$ to 50 producing values of $\hbar\Omega$ from $\hbar\Gamma$ to $7\hbar\Gamma$, respectively. To reach R_C the atom pairs must have a kinetic energy of $\hbar\Omega/2$, which is of the order of the Doppler temperature $k_\mathrm{B}T_\mathrm{D} = \hbar\Gamma/2$ ($T_\mathrm{D} = 125\ \mu$K for Cs). Thus one expects a strong temperature dependence. Based on the Landau–Zener model we have calculated the total suppression ratio by integration over the Maxwell–Boltzmann velocity distribution. To a first approximation we find a simple scaling law for the probability of ground-state collisions with the repump laser intensity, $\mathrm{P_{HCC}} \approx \exp(-s_0/A(T))$ [10]. The decay constant $A(T)$ varies approximately as $A(T) = 1 + 0.5(T/T_\mathrm{D})^2$. For simplicity we used only one repulsive molecular state with $V_{\mathrm{S+P}}(R) = +C_3/R^3$ with $C_3 = 12$ au [16]. However, we note that the result of our model changes very little if C_3 is changed by a factor of 2. The repump laser does not influence the light-induced collisions which appear therefore as an intensity-independent offset. We see this in the RE events resulting in one-atom losses which are shown in figure 4, (*c*) as squares.

To test our assumptions, we varied the MOT temperature by changing the cooling laser parameters (figure 4) and found significantly different decay constants as expected, $A(T) = 4.2 \pm 0.8, 9.2 \pm 2.7, 16.9 \pm 4.2$ for (*a*)–(*c*), respectively. From the measured decay constants we can directly infer the temperatures T, see figure 4. These results are in excellent agreement with previous temperature measurements [4, 6]. Extrapolating the curves to zero

repump intensity and taking into account the measured trap volumes we infer values for the total collisional rate coefficients $\beta_{\mathrm{HCC}} = (3.3 \pm 1.8), (4.6 \pm 1.7), (4.3 \pm 1.9) \times 10^{-11}$ cm^3 s^{-1} for (*a*)–(*c*), respectively. Here the uncertainty in the trap size of $\Delta r_0 \approx 2$ μm for all measurements makes the main contribution to the errors in β (typical relative errors of the non-normalized loss coefficients lie below 5% for $\beta_{2\mathrm{atoms}}/V$ and 20% for $\beta_{1\mathrm{atom}}/V$ and about 50% for absolute β values). Note that although the absolute values for loss event rates and for atomic densities differ substantially for different temperatures in figure 4, all calculated β_{HCC} are equal as expected for collisions between ground-state atoms. So we derive a final value for $\beta_{\mathrm{HCC}} = (4.1 \pm 1.0) \times 10^{-11}$ cm^3 s^{-1}. The value of β_{HCC} obtained from the measurement in figure 3 is now reasonably explained by optical suppression corresponding to a repump intensity of $s_0 = 4$ used for all data.

In summary, it has been shown that cold collision investigations on single trapped atoms provide novel and detailed information on the trap loss processes. We have measured the rate constant for the ground-state hyperfine-changing collisions partially hidden by strong optical shielding in previous studies. This intrinsic effect is always present in an alkali MOT. The method can also remove eventual ambiguities in experiments where an extra probe laser is introduced [9] in order to 'catalyse' different collisional loss channels. Note that such laser fields can also strongly affect the performance of the trap [17], and it is in general difficult to clearly discriminate between changed excitation conditions and changes in the atom number, both modifying the total fluorescence signal. It is also easy to generalize the method for studies of heteronuclear collisions [18] where fluorescence from different species can easily be spectrally distinguished. One can furthermore speculate about the possibility of watching the formation of an individual molecule from two atoms.

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