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Observation of flux enhancement in collisions between ultracold atoms

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We have observed a significant cooperative effect in ultracold trap loss collisions induced by two separate lasers. One laser, tuned close to the atomic resonance, excites the atom pair to an attractive potential at long range. The resulting acceleration and deflection give rise to an enhancement in the collisional flux of ground-state atoms reaching short range, as probed by a second laser. Enhancements up to a factor of ~ 3 have been observed. This indicates that the atomic pair-distribution function can be significantly distorted in a typical laser trap environment. [S1050-2947(96)51311-9]

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Recent experiments in ultracold collisions have revealed a variety of novel effects [1–3]. At these extremely low temperatures (e.g., $T<1$ mK), the atomic motion can be significantly affected by the long-range interatomic potentials. For example, in a collision between a ground-state atom and a laser-excited atom, the energy associated with the resonant dipole interaction can overwhelm the thermal energy, even for internuclear separations $R>100$ nm. This high ratio of potential to kinetic energy has been used successfully to suppress inelastic collisions by exciting the atom pair to a repulsive potential [4–8]. This shielding prevents the atoms from approaching closely enough for the inelastic process to occur. In the present work, we demonstrate the opposite process: long-range laser excitation of an atom pair to an attractive potential, which results in a significant (up to a factor of ~ 3) enhancement in the collisional flux at short range, as probed by a separate laser. In general, experimental measurements of ultracold collisional processes (e.g., photoassociation) are sensitive to the atomic pair-distribution function and, if performed in the environment of a typical laser trap, may be influenced by this flux enhancement.

The salient features of ultracold laser-induced trap loss collisions can be understood from the Gallagher-Pritchard model [9,10]. As two ground-state atoms approach in the presence of laser light (frequency ω) tuned below the atomic resonance (frequency ω_0), the atom pair can be excited to an attractive molecular potential as shown in Fig. 1. If the laser detuning $\Delta=\omega-\omega_0$ is negative, the attractive dipole-dipole potential $U=-C_3R^{-3}$ is resonant at the Condon point $R=(-C_3/\hbar\Delta)^{1/3}$. Once excited, the initially slow atoms accelerate towards each other, gaining kinetic energy. Since the excited state has a finite lifetime, it can decay before the atoms reach short range, effectively terminating the collision. This survival of the excitation from long range, where it is created, to short range, where an inelastic process leading to trap loss (e.g., a fine-structure change) occurs, is a crucial factor in the rate of trap loss collisions. Obviously, larger detunings result in better survival because the excitation occurs at smaller R where the acceleration is larger. However, there are other competing factors. At large detunings (small R) the number of available atom pairs (in a spherical shell of thickness dR) is significantly reduced by the phase-space factor $4\pi R^2dR$. Also, because the potential becomes steeper at smaller R, an atom pair passes through the Condon point more quickly, resulting in reduced excitation. Therefore, at small detunings (large R) there are many initially excited atom pairs, but their survival to short range is poor, while at large detunings (small R) there are fewer excited pairs, but their survival is greatly improved.

In the present work, we demonstrate a cooperative effect between collisions induced by two lasers, as shown in Fig. 1. The first laser (e.g., the trap laser), tuned close to the atomic resonance, excites a large number of atom pairs and causes significant acceleration and deflection before spontaneous emission intervenes. The second (probe) laser, tuned well below the atomic resonance, intercepts this enhanced flux and reexcites it, resulting in a short-range inelastic trap loss process. With only the first laser present, significant flux reaches short range, but the atoms arrive predominantly in the ground state, so trap loss does not occur. If only the second laser is present, less flux arrives at short range, but the excited fraction is higher. The signature of the cooperative effect is a collisional loss rate with both lasers present

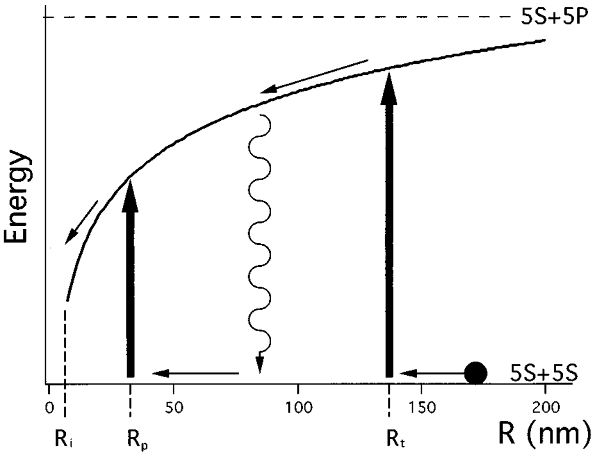

FIG. 1. Schematic of the flux enhancement effect. The atom pair approaches on the ground-state potential and is first excited by the trap laser at R_t. Acceleration and deflection occur before spontaneous emission returns the atom pair to the ground state. Reexcitation by the probe laser takes place at R_p followed by further acceleration. Excited atoms that reach the inelastic radius R_i result in trap loss.

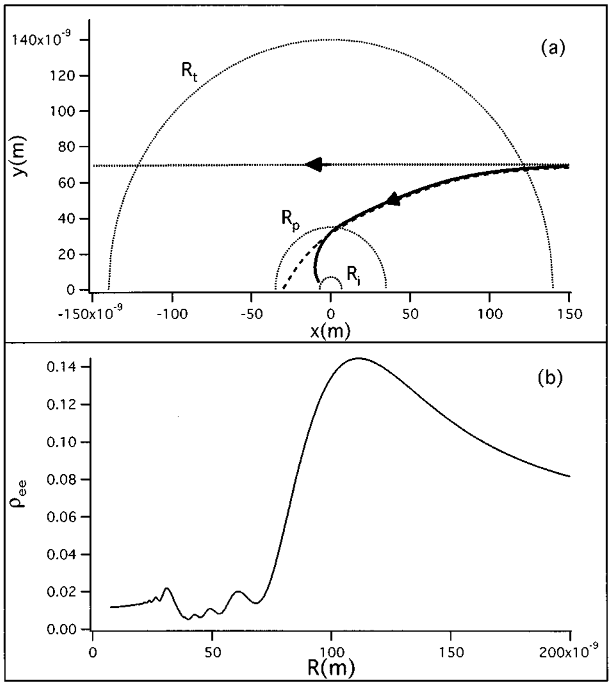

FIG. 2. (a) Atomic trajectories in the presence of the trap laser alone (dashed line), the probe laser alone (dotted line), and trap and probe lasers together (solid line). The circles indicate the Condon radii $R_t = 140$ nm and $R_p = 35$ nm, where the trap and probe lasers are resonant with the attractive potential, and the inelastic radius $R_i = 7$ nm. (b) ρ_{ee} as a function of R for the solid curve in (a). The transit times for $R_t \rightarrow R_p$ and $R_p \rightarrow R_i$ are 293 and 34 ns, respectively. Parameters are: $I_t = 3.8$ mW/cm^2, $\Delta_t = -\Gamma_A = -2\pi(5.9$ MHz), $I_p = 7.6$ mW/cm^2, $\Delta_p = -2\pi(400$ MHz), $v_0 = 15$ cm/s, $b = 70$ nm.

that exceeds the sum of the loss rates due to each laser individually. This effect is a purely mechanical one due to an enhanced flux of ground-state atoms. It is not a stepwise excitation requiring the presence of both lasers.

In order to gauge the magnitude of the cooperative effect, we have performed semiclassical simulations based on the optical Bloch equations [11] (OBE's). Three situations are compared. In the first, only the trap laser [$|\Delta_t| \sim \Gamma_A$ where $\Gamma_A = 2\pi(5.89$ MHz) is the atomic decay rate] is present; in the second, only the "probe" laser ($|\Delta_p| \gg \Gamma_A$) is present; and in the third, both lasers are present. A trajectory with initial relative velocity v_0 and impact parameter b is followed in the presence of excitation by the laser(s) to a single attractive potential. This potential is taken to be the 0_u^+, which has $C_3 = 1.1 \times 10^{-47}$ J m^3 and a decay rate $\Gamma = 4\Gamma_A/3$ (Ref. [10]). Each Rabi rate is taken to be $\Omega = (3)^{-1/2}\Omega_A$, where $\Omega_A = \Gamma_A[I/(3.24 \text{ mW/cm}^2)]^{1/2}$ is the atomic Rabi rate and I is the laser intensity. The factor of $(3)^{-1/2}$ is a result of directional averaging of the collision axis relative to the polarization axis [12]. The internal state of the atom evolves according to the OBE's, while its trajectory is determined by the attractive force weighted by the local excited-state fraction ρ_{ee}.

In Fig. 2(a) we compare sample trajectories for the three cases: trap laser only, probe laser only, and both lasers together. The impact parameter chosen clearly displays the cooperative effect. Only in the presence of both lasers does the atom reach $R_i = 7$ nm, the radius at which we assume the inelastic process (involving an excited atom) occurs. In Fig. 2(b) ρ_{ee} is plotted as a function of R for the case of both lasers present. When trajectories over a wide range of impact parameters are analyzed, the general features discussed above become evident. With the trap laser only, a large number of trajectories are initially excited near R_t and experience significant acceleration and deflection. However, their excitation decays away before reaching short range [13]. With the probe laser only, a smaller number of trajectories are affected, but survival of their excitation (created near R_p) is greatly improved. With both lasers present, there are several trajectories [e.g., Fig. 2(a)], which would not have yielded excitation at short range in the presence of either laser alone. This is the signature of the cooperative effect: enhanced flux caused by trap laser excitation is intercepted and guided to short range by the probe laser. In terms of angular momentum, we can think of the flux enhancement as helping the atom pair overcome the long-range centrifugal barrier, thereby allowing more partial waves to contribute.

In order to quantify the effect, we follow a large number of trajectories, labeled by j, whose impact parameters are separated by $\delta b = 2.5$ nm. The contribution of the jth trajectory to the effective cross section σ is weighted by the corresponding value of ρ_{ee} at R_i. The trap loss collision rate constant is then defined as

$$\beta = v_0\sigma = v_0 \sum_j \delta\sigma_j = v_0 \sum_j \rho_{ee}^j(R_i) 2\pi b_j \delta b. \qquad (1)$$

Since we are interested in the additional probe-induced trap loss rate caused by trap laser flux enhancement, we define an enhancement factor η:

$$\eta = \frac{\beta_{t+p} - \beta_t}{\beta_p} = \frac{\beta_p'}{\beta_p}, \qquad (2)$$

where β_t, β_p, β_{t+p} are the loss rates for trap laser only, probe laser only, and trap and probe lasers together. Note that if the contributions to the trap loss rate from the trap and probe lasers are independent (i.e., no cooperative effect), then $\eta = 1$. For the case shown in Fig. 2 ($\Delta_p/2\pi = -400$ MHz), $\eta = 2.8$. For a larger probe detuning, $\Delta_p/2\pi = -1$ GHz, we find $\eta = 4.5$. This increase in η with $|\Delta_p|$ is due primarily to a reduced value of β_p at larger detunings.

The experiment is performed by comparing the collisional loss rate β for ^{85}Rb atoms when the trap and probe lasers are present either simultaneously or separately. The magneto-optical trap [14] (MOT) is loaded from a laser-slowed atomic beam. When the loading is turned off, the number of atoms in the trap (measured by their fluorescence) begins to decay, in part due to ultracold inelastic trap loss collisions that eject atoms at a rate (per atom) of βn. Analysis of the decay curve, together with absolute measurements of the trapped atom density n, determines β.

In order to compare loss rates for the two cases (trap and probe together vs trap and probe separately applied), we chop the two lasers with a 50% duty cycle at a frequency of 5 kHz. When the two lasers are chopped in phase, the trap and probe are present simultaneously, and the cooperative effect due to flux enhancement is expected. When the two lasers are chopped out of phase (i.e., alternated in time), their contributions to the loss rate will simply add. Since we make our comparison by changing only the phase of the chopping, systematic uncertainties (e.g., trap volume) in measuring β cancel when we look at the ratio.

The MOT is operated at a total trap intensity (sum of all six beams) $I_t = 3.8$ mW/cm^2 and a detuning $\Delta_t = -\Gamma_A = 2\pi(-5.9$ MHz) relative to the $5S_{1/2}(F=3) \rightarrow 5P_{3/2}(F'=4)$ transition at 780 nm. The spatially filtered diode laser beams are Gaussian with a $1/e^2$ diameter of 6.3 mm. A pair of coils with opposing currents produces an axial magnetic field gradient of 4.8 G/cm. From previous time-of-flight measurements [15], the temperature is known to be ~ 50 μK ($v_0 \sim 15$ cm/s) under these conditions. A repumping laser, which is on continuously, is tuned to the $5S_{1/2}(F=2) \rightarrow 5P_{3/2}(F'=3)$ transition and prevents optical pumping into the lower ($F=2$) hyperfine level of the ground state. These operating conditions of the trap are chosen in order to minimize the collisional loss rate β_t. The intensity is above that necessary to recapture products of ground-state hyperfine-changing (ΔF) collisions and low enough to minimize contributions from ground-excited collisions [16]. We note that the low-temperature suppression effect [17] is partially responsible for the low value of $\beta_t = 2.5 \times 10^{-13}$ cm^3 s^{-1}.

The probe laser is combined with the trap laser (before the spatial filter) and is therefore identical to it with regard to geometry, beam size, and polarization. The two lasers are chopped with acousto-optic modulators. We are careful to keep the probe laser detuning large enough $[|\Delta_p| > 2\pi(300$ MHz)] and its intensity low enough ($I_p \leqslant 8$ mW/cm^2) to avoid perturbing the trap. This is verified by measuring the volume (and position) of the trapped cloud, as well as its fluorescence, in the presence and absence of the probe laser.

The collisional loss rate is measured for three situations. First, the probe laser is blocked and the loss rate due to the (chopped) trap laser alone is measured. This yields β_t. Then both the trap and probe lasers are applied, but alternated in time. This yields $\beta_t + \beta_p$. Since the two lasers are temporally separated, the contribution due to the probe laser alone is $\beta_p = (\beta_t + \beta_p) - \beta_t$. Finally, both lasers are applied, chopped in phase in order to be present simultaneously. This yields β_{t+p}. The enhanced (by the trap laser) rate of collisions caused by the probe laser (not including those collisions caused by the trap itself) is $\beta'_p = \beta_{t+p} - \beta_t$. For a fixed probe detuning, we measure β_p and β'_p at various probe intensities and verify the expected linear dependence, an example of which is shown in Fig. 3. For this particular example, the enhancement factor η [Eq. (2)] is ~ 1.4.

We present η as a function of probe detuning Δ_p in Fig. 4. A maximum enhancement of $\eta \sim 3$ is observed at the largest detuning (1 GHz). We are unable to measure β_p reliably for $|\Delta_p|/2\pi > 1$ GHz because of our limited probe intensity. Although the trend of increasing η with increasing $|\Delta_p|$ is reproduced in the simulations, measured values are significantly smaller than results of the simulations. This is not surprising in light of the simplicity of our model (i.e., classical trajectory, single attractive potential, no hyperfine structure) and shortcomings of the OBE method at low energies due to ambiguities in the trajectories [18–20].

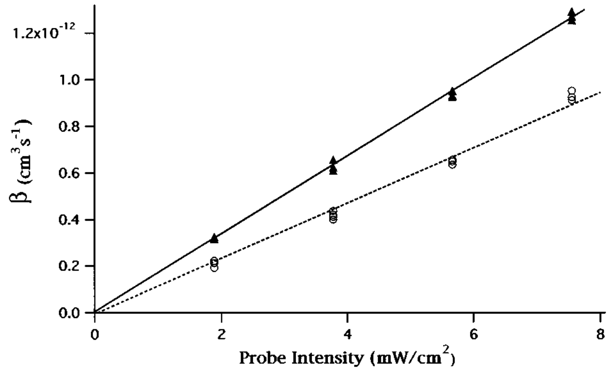

FIG. 3. Collisional loss rate β vs probe laser intensity I_p for $\Delta_p = -2\pi(400$ MHz). The open symbols indicate β_p, i.e., the trap and probe alternated out of phase, while the closed symbols indicate β'_p i.e., the trap and probe alternated in phase. The ratio of slopes of the best-fit lines given an enhancement factor $\eta = 1.4$.

The enhancement is independent of the chopping frequency over the range 1.7–100 kHz. Below 1.7 kHz, the enhancement factor begins to rise. We believe this is due to ballistic expansion (and therefore lower density) of the trapped cloud during the trap-off period. This is supported by the fact that the time-averaged volume of the trapped cloud also begins to increase at the lowest frequencies. We note that although the duration of an ultracold collision is relatively long [e.g., ~ 300 ns in Fig. 2(b)], our chopping frequency is always slow on this time scale.

In conclusion, we have observed a cooperative effect in trap loss collisions induced by two lasers, one tuned slightly below the atomic resonance (e.g., $\Delta_t = -\Gamma_A \rightarrow$ excitation occurs at $R_t = 140$ nm) and the other tuned much farther below resonance (e.g., $\Delta_p = -68\Gamma_A \rightarrow$ excitation occurs at $R_p = 35$ nm). The first excitation deflects and accelerates the incoming atom, but spontaneous decay (before an inelastic process can occur) results in a relatively small loss rate caused by this laser acting alone. However, its effect on the trajectories (deflection and acceleration) significantly enhances the flux of ground-state atoms that is available for excitation by the second laser. We have observed enhancements up to a factor of ~ 3. Semiclassical simulations predict the observed trend of larger enhancements as the detuning of the second (probe)

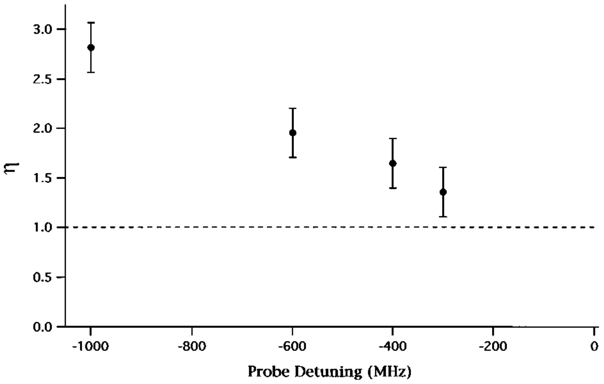

FIG. 4. Enhancement factor η vs probe laser detuning Δ_p. Error bars represent statistical uncertainties.

laser is increased. Our measurements indicate that these effects must be considered in any ultracold collision experiment using a separate probe (or ‘‘catalysis’’) laser to investigate the collision dynamics [21–25]. If the trapping laser is on simultaneously, its enhancement of the collisional flux may significantly alter the measurements. Since the enhanced flux resides in the ground-state, inelastic ground-state collisions (e.g., hyperfine-changing collisions [16] or ionizing collisions of metastable rare gases [6,7]) may be similarly affected. This point also applies to photoassociative spectroscopy experiments using ultracold atoms [26]. In this context, the effect may actually prove beneficial, resulting in increased rates of molecule formation at short range. Finally, we note that this flux enhancement induces nontrivial density correlations [9]. At high densities, such effects may become very significant.

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