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Time-Resolved Studies of Ultracold Ionizing Collisions

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Using 40 ns laser pulses, we probe the real-time dynamics of ultracold ionizing collisions in metastable xenon. We time resolve both shielding and enhancement effects, and observe the production of Xe$^2_2$ molecular ions through associative ionization. We estimate the rate of molecule formation in excited-state collisions, and directly measure the role of both flux enhancement and excited state survival in the collisional enhancement process. Conceptually simple theoretical models are used to predict the dynamics of the collisional shielding. [S0031-9007(98)06336-4]

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The study of ultracold ($T \leq 1$ mK) collisions between atoms has attracted a great deal of interest in recent years, in part because the long time scales involved ($t \sim 1 \mu s$) allow for absorption and emission of photons during the collision process. Such photon scattering can increase or decrease the collision rate, allowing control of the collision process by the application of appropriately tuned laser light [1–5]. The low velocities and long distances involved provide an opportunity to monitor atomic collisions in real time. Using short pulses of laser light, we probe the dynamics of the collisions on a $\sim 100$ ns time scale. We distinguish between Penning ionization ($\text{Xe}^+ + \text{Xe}^* \rightarrow \text{Xe}^{+} + \text{Xe} + e^-$) and the formation of $\text{Xe}_2^+$ molecules through associative ionization ($\text{Xe}^* + \text{Xe}^* \rightarrow \text{Xe}_2^+ + e^-$), and directly observe the role of excited-state survival and flux enhancement effects in collision enhancement. We also time resolve the collision shielding process, and use a simple theoretical model to predict the time behavior of this process. We note that Gensemer and Gould [6] have recently studied the time dependence of trap-loss collisions in a rubidium magneto-optic trap (MOT).

The basic physical picture of laser modification of collisions is shown in Fig. 1 and presented in detail in Ref. [7]. A pair of two-level atoms approaching each other in their ground ($S$) states (the “ground state” here being a metastable state with enough internal energy for Penning ionization) absorbs a photon from a laser tuned near the $S \rightarrow P$ transition. The laser excites the atoms to one of the two $S + P$ molecular excited state potentials \[U(R) = \pm \text{C}_3^2/R^3\] at a specific internuclear separation \[R_c = (\text{C}_3/\hbar \Delta)^{1/3}\] (the Condon radius), determined by the detuning \[\Delta = (\omega - \omega_o)\] from atomic resonance, and the long-range dipole-interaction strength \text{C}_3. Typical experimental values of $R_c$ are $\sim 1000 a_o$, (where $a_o$ is the Bohr radius) well outside the region of small internuclear separation where inelastic processes such as Penning or associative ionization occur. For blue-detuned ($\Delta > 0$) light, the atom pair is excited to the repulsive molecular state, and is prevented from reaching the inelastic collision region. This optical shielding suppresses the collision rate.

For red-detuned ($\Delta < 0$) light, the pair is excited to the attractive molecular state, and the atoms are accelerated towards one another. We then find an enhancement of the Penning ionization rate from one of two processes. Some fraction of the atom pairs thus excited will survive in the excited state long enough to reach the inner region, producing excited-state Penning ionizing collisions (EPI), which would not occur in the absence of the laser light. The remaining pairs will spontaneously decay back to the ground state before an inelastic collision occurs. They may, however, gain enough energy from acceleration on the excited potential to overcome centrifugal barriers and collide in higher angular momentum channels than their thermal energy would normally allow; this gives an enhancement of the

FIG. 1. Schematic of the laser-modified collision process. (a) Shielding ($\Delta > 0$): Atoms approach on the ground $S + S$ potential and are excited by the probe laser to the repulsive $S + P$ potential. They are reflected without reaching the short-range ionizing collision region. (b) Enhancement ($\Delta < 0$): Atoms are excited to attractive molecular states, and accelerated together. After some time, they may decay to the ground state and continue in at higher velocity (flux enhancement), or remain on the excited state into the ionization region.
ground-state Penning ionization (GPI) rate. This is the “flux enhancement” effect of Ref. [3], and is most important for extremely long-range excitations, where the probability of survival is low.

These processes may also enhance the rate of associative ionization [8]. The rate of ground-state associatively ionizing collisions can be increased through flux enhancement, and pairs surviving in the excited state can undergo excited-state associative ionization (EAI), colliding to form molecules. The doubly excited (P + P) potential is flat at long range, and should produce no enhancement of either rate for those few pairs excited to it.

The real Xe+ situation is more complex than this two-level model. There are 20 attractive and 20 repulsive potentials stemming from the 6s[3/2] + 6p[5/2]; separated-atom asymptote, all of which may contribute to the shielding or enhancement. These states arise only from different arrangements of molecular orbitals, and not from atomic hyperfine structure [9]. This is a significant simplification, as there are no crossings between the potentials, and we view the Xe+ situation as an ensemble of many two-level systems, each undergoing laser excitation at a different Rc.

The present experiment consists of applying a short (≤40 ns rms width) pulse of laser light tunered near the 882 nm 6s[3/2] → 6p[5/2] transition to an ultracold sample of atoms in a Xe+ MOT. The apparatus for cooling and trapping metastable xenon is described in Ref. [10]. The slowing and trapping laser beams are chopped at a rate of ~2 kHz, with an “off” period of 80 µs. Another laser is used to generate the probe pulse 40 µs after the trapping lasers are extinguished. The collision rate is observed by monitoring the rate of ion production. Ions are drawn into and detected by a channel electron multiplier mounted ~6 cm away, and the ion counts are recorded with a multichannel scaler, providing a histogram of ion production vs time.

The probe laser excites those atom pairs with internuclear separations near Rc, which then undergo either an enhancement or shielding process, depending upon the probe detuning, giving rise to a brief increase or decrease in the collision rate. Atom pairs at radii far from Rc are unaffected, and provide a constant background of ionizing collisions.

Typical data for both red and blue detunings are shown in Fig. 2. The ~750 mW/cm² probe pulse [11] is applied at t = 0, and the ~9 µs delay before observation of the enhancement or suppression effect reflects the time of flight to our detector. We measure this time of flight in a separate experiment (inset) using the signal from direct photoionization of Xe+ atoms in the 6p state in the MOT by a 5 ns pulse from a 514 nm dye laser. This signal, peaked at 8.90(5) µs [12], also shows the resolution of our ion collection system, with an rms width of 110 ns; convolved with the 40 ns pulse width, this gives us an experimental resolution of ~120 ns.

Figure 2(a) presents data for red (∆ < 0) detunings. The initial large enhancement peak is due to Xe+ ions, from Penning ionization in either flux-enhanced 6s + 6s (GPI) or 6s + 6p (EPI) collisions. The second peak is from Xe+2 molecular ions formed through associative ionization. Figure 2(b) presents data for blue (Δ > 0) detunings of the probe laser, showing the shielding effect. Collision suppression is seen at the time when the atoms excited at Rc by the probe laser would have collided had they continued to approach each other with their normal thermal velocity. The peak enhancement for red detunings occurs earlier than the minimum in the shielding data, showing the acceleration in the enhancement processes.

The associative ionization feature seen in the enhancement data is noticeably absent in the shielding case. As shielding affects only 6s + 6s collisions, this indicates that the formation of molecular ions is due only to atom pairs which survive in and collide on the excited-state potentials (EAI). We thus observe the excited-state survival which has long been thought to play an important role in the enhancement of ultracold collisions [13–15]. Excitestate survival in this system does not involve the population of long-lived states (such as the 1u and 2u states in alkali systems), which have been considered in previous work [6,14,15]. There is no attractive molecular potential from the 6s + 6p asymptote with a lifetime more than 20% longer than the 34 ns atomic lifetime [16]. The states in Xe+ analogous to the long-lived alkali states are repulsive, and do not contribute to the enhancement effect.

FIG. 2. Time-resolved collision signals from a ~750 mW/cm², 40 ns probe pulse at t = 0. (a) Collision enhancement for ∆ < 0 (Γ = 5 MHz). The first peak is from Xe+ (Penning ionization), the second, a factor of √2 later in time, from Xe+2 (associative ionization). Inset: Expanded view of the full time of flight signal, including the signal from direct photoionization of the MOT in a separate experiment (solid line), peaked at τc = 8.90(5) µs. (b) Shielding for ∆ > 0. The solid line is calculated for ∆ = 4Γ using the model discussed in the text.

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As the probe is detuned farther to the red [Fig. 2(a)], the magnitude of the enhancement decreases, in a manner consistent with steady-state experiments [1]. We find that the ratio of Xe$^+$ counts [$N_{PI}(\Delta) = N_{EPI}(\Delta) + N_{GPI}(\Delta)$] to Xe$^+$ counts [$N_{AI}(\Delta) = N_{EAI}(\Delta)$ only] decreases from greater than 10:1 at the smallest detunings used, approaching 4:1 at large $\Delta$ (Fig. 3). As we excite at larger $\Delta$ (smaller $R_c$), the survival probability for atom pairs excited to the $6s + 6p$ potential should approach 100%, as the time required to travel from $R_c$ to small $R$ decreases.

The Penning ion counts are then predominantly due to collisions occurring in the excited state, calculated using theoretical values of $C_3$ [16]. We then calculate the distribution of collision times resulting from these populations, summing the $s$, $p$, and $d$ partial waves ($l = 0, 1, 2$) [18]. We average the resulting curves, weighted by $R_c^2$ to account for the number of atoms in a shell of radius $R_c$, and convolve the result with our experimental resolution.

The solid curve in Fig. 2(b) shows the result of such a calculation for $\Delta = 4\Gamma$ (where $\Gamma = 5$ MHz is the natural linewidth), scaled to match the amplitude of the experimental signal. The general shape of the curve is well reproduced by the theory, though the theoretical curve is somewhat narrower than the data. It may be possible to improve the agreement by including excitation over a range about $R_c$, rather than the $\delta$-function excitation assumed here [7].

Figure 4 shows the measured collision time (referred to the ion time of flight signal shown in Fig. 2) as a function of $1/\Delta^{1/3}$ (which is proportional to $R_c$). The success of this simple model of shielding (solid line) is striking. The data fit well to a line, consistent with our picture of atoms moving together at constant velocity from the Condon radius. Comparing the slope of the fit to slopes calculated using single values of $C_3$ [19], we extract an effective $C_3$ for the shielding process, using the thermal velocity of

![Graph](https://example.com/graph.png)

**FIG. 3.** The ratio of total counts in the Penning ion signal ($N_{PI}$) to total counts in the associative ionization signal ($N_{AI}$), as a function of detuning.

![Graph](https://example.com/graph.png)

**FIG. 4.** Collision times $\tau$ as a function of $(1/\Delta)^{1/3}$ ($\propto R_c$). Shielding and Penning ionization times are measured from the peak of the ion time of flight signal ($t_o$) shown in the inset in Fig. 2 to the point of minimum or maximum collision rate. Associative ionization collision times are referenced to $\sqrt{2} t_o$. The error bars reflect systematic uncertainties in the measurement of peak positions. The dashed line is a fit to the associative ionization data using the survival time of Ref. [15]; the $\tau = -40$ ns intercept is consistent with $\tau = 0$. The solid line is the prediction of the simple model discussed in the text.
$v_T = 6.0 \text{ cm/s}$, and find a value of $11(3) \times (e a_0)^2$ [12] ($e$ is the electron charge). This is in agreement with the $11.0(e a_0)^2$ average (using the same $R^2$ weighting function) of the theoretical $C_3$ values. By changing the detuning of the trapping lasers, we varied the thermal velocity by a factor of 2, and find that the collision times are linear in $1/v_T$, again in agreement with our model.

Also shown in Fig. 4 are the observed collision times for enhancement of both Penning and associative ionization. The broken line is a fit to the associative ionization data using the collision time calculated from the simple model in Ref. [15]. Collision times for both processes are significantly shorter than for the shielding case, showing that acceleration on the excited-state potential is the dominant effect in determining the collision time. This is supported by the observation that both Penning and associative collision times are independent of the initial velocity $v_T$.

In conclusion, we use short laser pulses to study the time dependence of ionizing collision processes in detail. We observe the production of molecular ions from excited-state collisions, and estimate the rate of molecule formation from such collisions. We show that the flux enhancement effect dominates the collision enhancement process for long-range excitations, and must be included in models of collisions in optical traps. We find that conceptually simple models provide good qualitative and quantitative agreement with the data. Interesting theoretical issues remain in interpreting the details of the collision enhancement and associative ionization processes.

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[8] The relative rates of Penning and associative ionization will depend upon the molecular potentials at short range, which have not been calculated for this system.
[9] Unlike alkali systems, the $^{132}$Xe isotope used for this work has no nuclear spin.
[11] Studies of the intensity dependence show that both shielding and enhancement processes begin to saturate at intensities of $\sim 400 \text{ mW/cm}^2$. This is consistent with the theory of Ref. [7].
[12] All quoted uncertainties are 1-standard deviation values.
[18] The $d$-wave contribution is quite small, and higher partial waves are negligible.
[19] Numerically, we find that for distributions calculated using single values of $C_3$, the slopes fit to plots like Fig. 4 must be multiplied by a factor of 2.8 to extract the original $C_3$. We apply the same correction to the fit to our data.