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Observation of Cold Collisions between Trapped Ions and Trapped Atoms

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We study cold collisions between trapped ions and trapped atoms in the semiclassical (Langevin) regime. Using Yb⁺ ions confined in a Paul trap and Yb atoms in a magneto-optical trap, we investigate charge-exchange collisions of several isotopes over three decades of collision energies down to 3 μeV \( (k_B \times 35 \text{ mK}) \). The minimum measured rate coefficient of \( 6 \times 10^{-10} \text{ cm}^3 \text{s}^{-1} \) is in good agreement with that derived from a Langevin model for an atomic polarizability of 143 a.u.

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Studies of cold collisions between trapped neutral atoms have revealed a plethora of fascinating quantum phenomena, including Wigner threshold laws [1], magnetically tunable Feshbach resonances [2], controlled molecule formation [3], and the suppression of individual scattering channels [4]. Collisions between trapped ions, on the other hand, are featureless since the strong long-range repulsive Coulomb interaction prevents the ions from approaching each other. Collisions between ions and neutral atoms [5–8] fall into an intermediate regime where an attractive long-range \( r^{-4} \) potential leads to semiclassical behavior for a wide range of collision energies, but where quantum phenomena dominate at very low energies. Cold ion-atom collisions have been proposed as a means to implement quantum gates [9], to cool atoms [7,10] or molecules [11,12] lacking closed optical transitions, to bind small Bose-Einstein condensates to an ion [13], or to demonstrate novel charge-transport dynamics [14].

As a function of collision energy, charge-exchange and momentum-transfer ion-atom collisions exhibit three distinct regimes [6,7]: a high-energy classical (hot) regime with a logarithmic dependence of cross section \( \sigma \) on energy \( E \), a wide semiclassical Langevin (cold) regime with a power-law dependence \( \sigma(E) \propto E^{-1/2} \) [5,6], and a quantum (ultracold) regime where contributions from individual partial waves can be distinguished. However, given the large forces on ions produced by small stray electric fields, it has been difficult to reach experimentally even the semiclassical regime. References [15,16], studying charge exchange at \( E \sim 100 \text{ meV} \), report the only observations of Langevin-type ion-atom collisions. In ion-molecule systems, experimental signatures of Langevin collisions have been seen at high temperature [17–19], and recently also at 1 K (80 μeV) [20]. In all previous work, at most one of the collision partners was trapped.

In this Letter, we study collisions between independently trapped, laser-cooled ions and atoms down to unprecedented low energy (3 μeV) in the semiclassical collision regime. Using a double-trap system [21], we investigate resonant charge-exchange collisions for different Yb⁺ + Yb isotope combinations and find agreement with the Langevin model to within a factor of 2 over three decades of energy [5,6]. The highest energy, 4 meV = \( k_B \times 45 \text{ K} \), corresponds to the transition to the classical regime [22,23], while at the lowest energy, 3 μeV = \( k_B \times 35 \text{ mK} \), where approximately 40 partial waves contribute to the cross section, isotope shifts should become relevant. The lower limit on collision energy is set by our ability to detect and minimize the micromotion of a single ion in the Paul trap.

The long-range interaction potential between a singly charged ion and a neutral atom is the energy of the induced atomic dipole in the ion’s electric field, given by \( V(r) = -C_4/(2r^6) \), where \( C_4 = aq^2/(4\pi\epsilon_0)^2 \) is proportional to the atomic polarizability \( \alpha \), and \( q \) is the electron charge. For a given collision energy \( E \) in the center-of-mass frame, there exists a critical impact parameter \( b_c = (2C_4/E)^{1/4} \) that separates two types of collisions: those with impact parameter \( b < b_c \) that result in inward-spiraling orbits of radius approaching zero, and those with \( b > b_c \) that never cross the angular-momentum barrier [5,24].

For collisions of an ion with its parent atom, a semiclassical resonant charge-exchange cross section \( \sigma_{ce} \) can be simply derived, provided that \( b_c \) is large compared to the range of the molecular potential. For collisions with \( b > b_c \), the electron should remain bound to the incoming atom. For close-range collisions with \( b < b_c \), the electron in the resonant process \( A^+ + A \rightarrow A + A^+ \) is equally likely to exit attached to either nucleus, resulting in either a charge-exchange collision or an elastic collision. It follows that the resonant charge-exchange cross section is \( \sigma_{ce} = \sigma_L/2 \), where \( \sigma_L = \pi b_c^2 \) is the Langevin cross section. The corresponding semiclassical rate coefficient \( K_{ce} = \sigma_{ce} \nu = \pi \sqrt{C_4}/\mu \), where \( \nu \) is the relative velocity and \( \mu \) the reduced mass, is independent of energy.

At high energies this model becomes invalid when \( b_c \) becomes so small that charge exchange outside the centrifugal barrier starts to play a role; for Yb, this occurs at energies \( E \gtrsim 10 \text{ meV} = k_B \times 120 \text{ K} \) [6,22,23]. At very

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low energies, the semiclassical Langevin model ceases to be valid when the s-wave scattering limit is reached, which happens near $E_s \sim h^4/(2\mu^2C_4) = 4 \text{ peV} = k_B \times 50 \text{ nK}$. Quantum simulations [6] of this system [22] confirm the semiclassical Langevin model in the indicated energy range. For collisions between different isotopes, the Langevin expression should be modified at low collision energies comparable to the small difference in binding energies of the electron to the two nuclei, i.e., the isotope shift of the ionization potential. In this case, we expect endoenergetic charge-exchange collisions to be suppressed [25,26].

We use a magneto-optical trap (MOT) in combination with a Paul trap, as originally proposed by Smith [11]. The setup, shown in Fig. 1, is described in detail elsewhere [21]. In the present work, $^{172}\text{Yb}$, $^{174}\text{Yb}$, or $^{171}\text{Yb}$ atoms are selectively loaded from an atomic beam into a MOT by tuning the laser frequency near the $^5S_0 \rightarrow ^1P_1$ transition at $\lambda_d = 398.8 \text{ nm}$. Typically, the MOT is operated at a 75 G/cm magnetic-field gradient and contains $3 \times 10^5$ atoms at a peak density of $2 \times 10^8 \text{ cm}^{-3}$ and a temperature of 700 $\mu\text{K}$ as determined by a time-of-flight measurement.

Cold ions are produced by nonresonant photoionization from the excited $^1P_1$ state of the MOT with 370-nm light from a semiconductor laser [21]. The ion trap is a surface-electrode Paul trap printed on a vacuum-compatible substrate [21], and is typically operated at 1.4 MHz to create a 0.3 eV deep pseudopotential trap 3.6 mm above the trap surface with a secular frequency of 67 kHz. Ion trap populations can be adjusted between a single and surface with a secular frequency of 67 kHz. Ion trap

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to the collision energy is negligible. For all energies investigated here, the atoms’ contribution to the collision energy is negligible. The other data points show observable Doppler broadening and were measured with up to a few thousand ions, resulting in smaller statistical uncertainties. We estimate systematic uncertainties to be a factor of 2 in $K$ due to the difficulty of absolute MOT density calibration, and $\pm 50\%$ in $E$, due to the nonthermal energy distribution of the micromotion.

The ab initio calculated value for the polarizability $\alpha_s = 143$ a.u. = $\epsilon_0 \times 2.66 \times 10^{-28}$ m$^3$ [22] for the atomic $^1S_0$ ground state yields $K_{ce} = 5.8 \times 10^{-10}$ cm$^3$ s$^{-1}$ for ground-state collisions (Fig. 4 solid line). Our experimentally measured value of $K = 6 \times 10^{-10}$ cm$^3$ s$^{-1}$ in the semiclassical region around $E \sim 100$ $\mu$eV is thus in good agreement with the Langevin model. The measured $K$ at the highest average collision energy $E_0 = 4$ $\mu$eV, close to the transition to the classical scattering region, is somewhat larger than the classical prediction [22,23]. Given the rapid Doppler cooling of the ions, the discrepancy is probably not due to collision-induced heating and trap loss. Rather, we speculate that it may be caused by averaging our nonthermal (micromotion) energy distribution over an oscillating cross section in the transition region [30]. Doubly excited collisions Yb$^{++} +$ Yb$^+$ cannot occur as the excitation light for ions and atoms is modulated out of phase. For Yb$^{++} +$ Yb collisions, the Langevin cross section, depending only on the ion’s charge and the atom’s polarizability, is unchanged, and the charge-exchange process remains resonant, yielding the same value of $\sigma_{ce}$. Yb$^+ +$ Yb$^+$ collisions cannot contribute for our collision energies since the atomic transition is tuned out of resonance with the MOT.
light at an ion-atom distance $R_0 = 30$ nm, larger than $b_c = 2\text{–}12$ nm, and modeling of collision trajectories shows that the time required to move between $r = R_0$ and $r = b_c$ exceeds several excited-state lifetimes. For illustration, we have indicated in Fig. 4 the small change in $K_{ee}$ if Yb$^+ + $Yb$^+$ collisions with $\alpha_{p} = 500$ a.u. for the $1P_1$ state [31] were to contribute at our measured MOT excited-state fraction.

The theoretical value $K_{ee} = \sigma_L \nu/2$, corresponding to equal binding probability of the electron to the two nuclei, should apply when the collision energy far exceeds the isotope shift of the ionization potential. The latter can be estimated from spectroscopic data of a transition to a state with low electron probability density at the nucleus, such as $4f^{14}6s^2 \rightarrow 4f^{14}6s10d$ [32]. It follows that the $^{172}$Yb$^+ + ^{174}$Yb $\rightarrow ^{172}$Yb$ + ^{174}$Yb$^+$ reaction should be exothermic and release $\Delta E = 2.9 \mu eV = h \times 0.7$ GHz. At the substantially larger collision energy $E = 0.21$ meV $\gg \Delta E$ we have investigated various isotope combinations, $^{172}$Yb$^+ + ^{174}$Yb, $^{174}$Yb$^+ + ^{172}$Yb, and $^{174}$Yb$^+ + ^{171}$Yb, and find that they all display the same $K$ (Fig. 4). We speculate that the data point $^{172}$Yb$^+ + ^{174}$Yb $\rightarrow ^{172}$Yb$ + ^{174}$Yb$^+$ at the lowest energy $E = 3.1 \mu eV$, corresponding to exothermic collisions with $E = \Delta E$, may exhibit an increased rate coefficient compared to $K_{ee}$.

While it may be impossible to compensate stray fields well enough to reach the $s$-wave scattering limit $E_s = 4$ peV [6,22], Feshbach and other collision resonances may be observable well above $E_s$ [8]. All the Yb isotopes used here have been cooled to quantum degeneracy in an optical dipole trap [33,34], which would allow the investigation of collision processes between an ion and a Bose-Einstein condensate or Fermi gas. Alternatively, to avoid the large resonant charge-exchange cross section observed here, a different species such as Rb could be used for sympathetic cooling of ions [7,10], or for studying ion impurities in a Bose-Einstein condensate [13].

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