Laser-Induced Ionizing Collisions of Ultracold Krypton Gas in the $1s_5$ Metastable State

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(Received 8 April 1994)

The line shape of laser-induced ionizing collisions of ultracold Kr gas is investigated near the $1s_5[4p^5 5s:{}^3P_2]$ and $2p_9[4p^5 5p:{}^3D_3]$ transition. The rate increases several times when the laser is red tuned by a few times of the natural width, while it sharply decreases for a slight blue detuning. This frequency dependence of the collision rate is due to the collision dynamics characteristic of ultracold atoms in a near-resonant light. Our experimental results on metastable Kr atoms show a good qualitative agreement with semiclassical Monte Carlo simulation of the cold collision dynamics.

PACS numbers: 34.50.Rk

Recent progress in laser cooling and trapping of neutral atoms has made it possible to study collisions at extremely low temperature. Using laser cooled atoms, collision rates with a well-defined atomic state, polarization, temperature, etc., have been measured by several groups [1–13]. One of the striking aspects of cold collisions in neutral atom traps is that the collision cross section changes dramatically with the application of a near resonant monochromatic light [6–13]. This change is caused by a long-range resonant dipole-dipole interaction that arises when one of the colliding atomic pair is excited to the upper state of the cooling transition. At ultracold temperatures, the dipole-dipole interaction can substantially alter the atomic velocities even at very large internuclear separation. An extremely large loss rate of the He* trap was explained by this mechanism [10]. In alkali traps, the inelastic collision rate was found to increase several times with the application of an auxiliary laser ("catalysis laser"), whose frequency was slightly below the frequency of the cooling transition [11–13]. Theory of the trap loss based on the dipole-dipole interaction was discussed in detail by Gallager and Pritchard [14]. Extended theoretical discussion on cold collisions was also given by Julienne *et al.* [15,16]. However, because of the existence of hyperfine levels and indistinguishable loss channels, quantitative comparison between the theory and the experimental result is difficult. Recent experimental result by Hoffmann *et al.* clearly show the necessity to include additional loss channels [12]. Furthermore, the time-consuming fluorescence decay measurements required in the alkali systems make it difficult to obtain reliable data close to a resonance, where the characteristic behavior of the cold collisions is most pronounced.

For a metastable rare-gas trap of even isotopes, the cooling transition has no hyperfine structure. Ionizing collisions are the dominant trap-decay mechanism and can be easily distinguished from other loss mechanisms. In this Letter we report experimental results on the light-induced ionizing collision rate of laser-cooled $1s_5[4p^5 5s:{}^3P_2]$ metastable Kr atoms. We detected ions instead of fluorescence. The advantage of ion detection was clearly shown in recent studies on cold Na collisions [1,4–6,9]. We observed not only the increase of the collision rate with a red tuned light, but also the decrease with a blue tuned light. Comparison of the experimental result with semiclassical Monte Carlo simulation shows a good qualitative agreement with the theory.

When a colliding pair of atoms is in two different internal states that are connected by a dipole transition moment, the two atoms are influenced by the resonant dipole-dipole interaction potential [17],

$$V(r) = \pm a\hbar\gamma(\lambda\!\!\!^{-}/r)^3, \tag{1}$$

with $\lambda\!\!\!^{-} = \lambda/2\pi$, where λ and γ are the wavelength and natural linewidth of the atomic transition, respectively, and a is a configuration dependent constant of the order of 1. If the two states have a large transition moment such as in the cooling transition of a neutral atom trap, the magnitude of this potential is much larger than the kinetic energy of the trapped atoms even at a distance of several tens of nanometers.

Suppose laser cooled Kr atomic gas in the $1s_5$ metastable state is illuminated by the catalysis laser slightly detuned from the cooling transition $1s_5$-$2p_9[4p^5 5p:{}^3D_3]$. Two approaching atoms will be excited to one of the $5 \times 7 \times 2$ adiabatic potential surfaces of the $1s_5$-$2p_9$ binary atomic system, at the distance at which the detuning of the laser becomes equal to the interaction energy, Eq. (1). If the laser is red tuned, the atomic pair moving along the $1s_5$-$2p_9$ potential surface feels an attractive force, and the atoms are accelerated towards a closer collision path. Therefore, the probability to cause inelastic collisions increases. The magnitude of the change varies, depending on the detuning. If the detuning is large, the pair is excited at the distance where the slope of the $1s_5$-$2p_9$ potential surface is steep. Therefore, the acceleration is large, but the excitation probability is small. If the detuning is less, the acceleration is smaller, but the excitation probability increases. In addition, the collision can take time longer than the natural lifetime $1/\gamma$, and the pair may return to the $1s_5$-$1s_5$ potential surface by emitting a spontaneous photon before it reaches the minimum distance. If the

0031-9007/94/73(19)/2555(4)$06.00

frequency is closer to the resonance, the pair repeats the excitation-deexcitation cycle before it becomes out of resonance. Therefore, the acceleration is much larger than that expected from the potential slope. When the laser is blue tuned, the pair is repelled, and the ionization probability decreases.

The consequence of the above physical picture is shown, in Fig. 1, as the result of the Monte Carlo simulation. The colliding atomic pair was assumed to take the classical trajectory determined by the potential surface of the pair and to be excited or deexcited instantaneously. We represented the $1s_5$-$2p_9$ potential surfaces with one attractive and one repulsive, each having the same slope $a = 0.64$ as in Eq. (1) that was the value averaged over all potential manifolds. We took the excitation rate to either of the surfaces as

$$\frac{\gamma}{4}\frac{I/I_0}{1 + I/I_0 + \{2[\Delta\nu - V(r)/\hbar]/\gamma'\}^2}, \qquad (2)$$

where $\Delta\nu$ was the detuning of the laser frequency from the $1s_5$-$2p_9$ atomic transition and I the power density of the laser. Here I_0 was the saturation power density corresponding to the transition that had the natural linewidth γ'. We assumed γ' was equal to that of the $1s_5$-$2p_9$ atomic transition, $\gamma = 1/(31\ \text{ns})$ [18,19]. The ionization probability of 1 was assumed when two atoms approached closer than the distance that was experimentally determined from the ionization collision cross section without the catalysis laser. The curves in Fig. 1 show the laser frequency dependence of the collision rate, normalized to the rate without the laser at the temperatures of 100 and 400 μK and the laser power of 5 mW/cm^2. The ionization radius of 13 and 8.9 nm were used at respective temperature. The rate was greater than 1 for red detuning and was smaller than 1 for blue detuning as expected. It changed rapidly from minimum to maximum when the laser was scanned through the resonance from blue side. At a larger red detuning, the rate gradually returned to 1 with the width of approximately 100 MHz. The simulation showed distinct three regions. At the temperature of 100 μK for the negative detuning beyond -80 MHz, the rate change was determined by the probability of excitation, and the pair remained in the $1s_5$-$2p_9$ state until it hit the minimum distance. For a smaller detuning, the collision took a longer time, and the probability of the pair in the $1s_5$-$2p_9$ state at the minimum distance decreased. Below -50 MHz most atoms decayed to the $1s_5$-$1s_5$ state. For the detuning less than -20 MHz, the multiple excitation cycle was set in making a sharp variation around the resonance. This was evident from the comparison at two different temperatures. At a higher temperature, the number of excitation cycles was less, and the net gain in the velocity was smaller. Therefore, the rate changed more moderately near the resonance. It should be noticed that the laser-induced ionizing rate was determined by the ratio between the amount of the acceleration and the velocity of the atom. Therefore, the peak height of the curves in Fig. 1 is inversely proportional to the square root of the temperature. This does not apply for alkalis, because in the vicinity of resonance, the atomic velocity is not an important factor to modify fine-structure changing collisions. The experimental results on Rb and Cs did not reveal temperature dependence, and the maximum was observed at the detuning more than 40 times larger than the natural width [11–13].

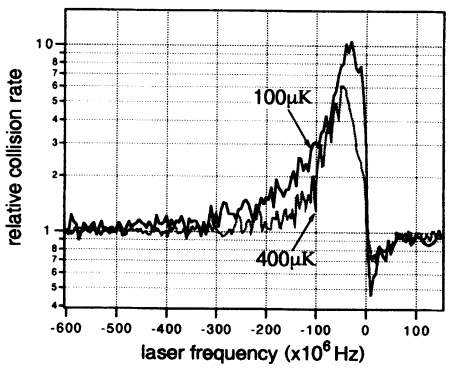

FIG. 1. The Monte Carlo simulation of the relative ionizing collision rate k_{cat}/k_{1s} as a function of the catalysis laser frequency.

The experimental procedure to measure light-induced ionizing collision rates of metastable Kr gas is simpler and more reliable, compared to the similar experiment on Rb and Cs [11–13]. We cool and trap Kr atoms under the vacuum pressure of approximately 10^{-7} Pa. At this pressure, the ions are produced only from inelastic collisions between two Kr atoms in the trap, and the ion count rate is proportional to the ionizing collision rate of the cold gas. Produced ions can be easily collected and counted by an electron multiplier placed at negative electric potential.

We used the standard magneto-optical trap [20] with three standing-wave lasers to cool and trap Kr, which was similar to the one used in our previous report [21]. One of the standing waves was parallel to the axis of the quadrupole magnetic field. The total intensity of the cooling and trapping lasers was 30 mW/cm^2 and their $1/e$ diameter 10 mm. The number of trapped atoms was typically 10^5 and the $1/e$ radius of the trap 300 μm. An electron multiplier (Hamamatsu R2362) was placed 5.8 cm below the trap, and its cathode was kept at the potential of -4.5 kV. It was used to count ions generated by the cold collisions and also to count metastable Kr atoms in the time-of-flight measurement to determine the temperature of the Kr cloud. The electron multiplier was equipped with a CuBeO cathode, and the quantum efficiency for metastable Kr atoms was measured to be 2.7%, in agreement with the reported value [22]. This low quantum efficiency for metastable atoms, together

with the timing described below, guaranteed us that the error coming from the metastable count was negligible in the ionizing collision measurement. A calibrated photomultiplier and a charge coupled device (CCD) camera were used to monitor the number of atoms in the trap and the trap size, respectively. The catalysis laser was introduced in the same path, with the same polarization and radius, as those of the two standing-wave cooling lasers perpendicular to the magnetic axis. The cooling lasers were periodically turned off with the width of 23 μs at the interval of 115 μs. The catalysis laser was turned on in every second period while the cooling laser was turned off. We measured the ion count rate approximately 10 μs after turning off the cooling laser for the duration of 8 μs. The time sequence of the measurement is shown in Fig. 2. When the cooling laser was turned off, the Kr atoms quickly decayed to the $1s_5$ metastable state, however, they could not move substantially during the 23 μs off period. Therefore, the atomic density did not substantially change during the switching cycle. The delay and duration of the ion counting were selected to avoid transient response of the collision rate, yet to minimize the disturbance of the catalysis laser on noncolliding atoms. The ratio of the ionizing collision rates with and without the catalysis laser k_{cat}/k_{1s} was obtained from the ratio of the ion count rates I_{cat}/I_{1s}.

The ionizing collisions of the laser cooled metastable Kr can have two channels: the Penning ionization collisions (PI),

$$\mathrm{Kr}(1s_5) + \mathrm{Kr}(1s_5) \rightarrow \mathrm{Kr}(^1S_0) + \mathrm{Kr}^+ + e,$$

and the associative ionization collision (AI),

$$\mathrm{Kr}(1s_5) + \mathrm{Kr}(1s_5) \rightarrow \mathrm{Kr}_2^+ + e.$$

In addition to the frequency dependence of the relative collision rate k_{cat}/k_{1s} we determined the absolute value of the collision rate k_{1s} and the ratio of the two channels $k_{1s}(\mathrm{PI})/k_{1s}(\mathrm{AI})$. To obtain k_{1s} we determined the atomic density n by measuring the fluorescence intensity and the size of the trap while the cooling laser was on. The collision rate k_{1s} is equal to I_{1s}/n^2V, divided by the quantum efficiency of the ion count, where V is the volume of the atomic cloud. Assuming the quantum efficiency of 50%, the result was $k_{1s} = 2 \times 10^{-10}\ \mathrm{cm}^3\mathrm{s}^{-1}$. The k_{1s} was almost constant in the temperature range between 500 and 50 μK. This value agreed within factor 2, with an independent measurement from the fluorescence decay of the trap. The result of cold-collision study from the trap decay will be presented elsewhere.

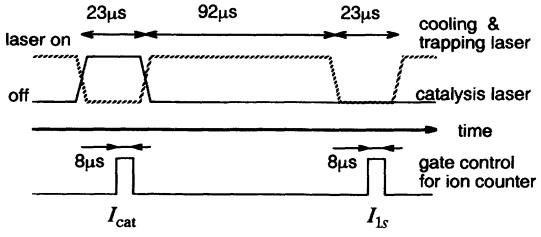

FIG. 2. The time sequence of the data acquisition.

The branching ratio $k_{1s}(\mathrm{PI})/k_{1s}(\mathrm{AI})$ was determined from the time-of-flight measurement of the transient ion signal when the intensity of the trapping laser was switched. It produced two peaks, separated by the ratio of $1:\sqrt{2}$ on the transit time of ions, as was expected from the mass ratio between Kr^+ and Kr_2^+. From the ratio of the two peaks, PI was found to be the dominant channel, $k_{1s}(\mathrm{PI})/k_{1s}(\mathrm{AI}) \approx 10$.

We measured the relative ionizing collision rate k_{cat}/k_{1s} as a function of the detuning of the catalysis laser and at various Kr temperatures. The temperature was controlled by changing the detuning of the cooling laser. It was measured without turning on the catalysis laser. Figure 3 shows k_{cat}/k_{1s} as a function of the detuning of the catalysis laser at the temperatures of 74 and 450 μK. The corresponding detunings of the cooling laser were -33 and -5 MHz, respectively. The total power density of the catalysis laser was 4 $\mathrm{mW/cm}^2$. Figure 4 shows k_{cat}/k_{1s} as a function of the intensity of the catalysis laser. The detunings of the catalysis laser were -30 and $+20$ MHz, and the temperature was 450 μK. In this power region both the peak and the dip in Fig. 3 increases almost linearly with the laser power. At the power above 10 $\mathrm{mW/cm}^2$, the trap shape was seen to change. However, the line shape of the frequency dependence of Fig. 3 was unaffected. The line shape of the experimental curves in Fig. 3 closely reproduces the theoretical result of Fig. 1. The reduction as well as the enhancement of the collision rate is observed. Both the position and the relative height of the peak and valley are in agreement. The effect of multiple absorption-emission cycle near the resonance is clear. The temperature dependence is also observed, though the change is somewhat smaller than expected from the simulation. The agreement is

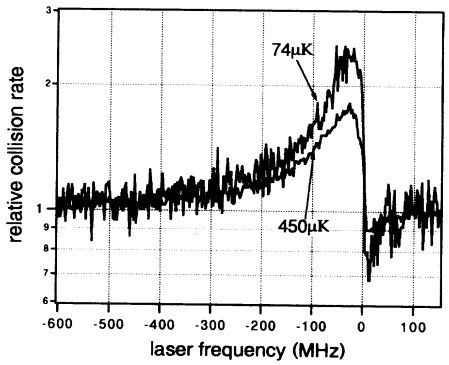

FIG. 3. The experimental result of the relative ionizing collision rate of Kr k_{cat}/k_{1s} as a function of the catalysis laser frequency. Two curves show the result at the temperatures of 74 and 450 μK.

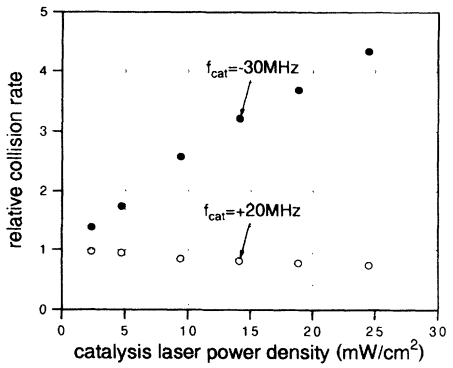

FIG. 4. The catalysis-laser intensity dependence of the relative ionizing collision rate k_{cat}/k_{1s}. The detunings of the catalysis laser were -30 MHz (filled circles) and $+20$ MHz (open circles) from the atomic $1s_5$-$2p_9$ resonance. The temperature was 450 μK.

unexpectedly good, considering that the simulation in Fig. 1 uses several simplified assumptions. The absolute change of the collision rate is approximately factor 3 smaller than the value in the simulation. Although the simulation shows a good agreement with the experiment close to the resonance, the calculated value of k_{cat}/k_{1s} drops off faster than the experimental value at the wing of the red detuning. The k_{cat}/k_{1s} will decrease more slowly if we use a larger γ' or a. However, its effect on the peak height is expected to be small. To obtain a better agreement, the entire $1s_5$-$2p_9$ potential surfaces and the variation of the transition moment with the atomic distance have to be properly taken into account in the simulation.

The clear cut result of the cold Kr ionizing collisions results from the possibility to study the process without the influence of the cooling laser and to distinguish various inelastic channels. Various other aspects of cold collisions of metastable rare gases can be studied with quality better than cold alkali atoms can do. Kr has an odd isotope ^{83}Kr with a nuclear spin of $\frac{9}{2}$. The hyperfine splittings of rare-gas atoms are much larger than those of alkali atoms, and each hyperfine level can be independently treated in the study of cold collisions. The comparison of ionization rate with ^{84}Kr using the catalysis laser may more clearly reveal the role of hyperfine interactions on the collision than the study of Rb and Cs. Metastable rare-gas atoms can be cooled below the temperature, at which the s-wave scattering becomes dominant. Comparison of various collision channels between even and odd isotopes will be also interesting. Detailed experimental work on the collision processes in metastable Kr and Ar traps is underway. The result will be given elsewhere.

This work was supported by a Grant-in-Aid from the Ministry of Education, Science and Culture.

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