Lifetime Measurement of the $1s_5$ Metastable State of Argon and Krypton with a Magneto-Optical Trap

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The natural decay rates of the $1s_5[np^5(n+1)s, {}^3P_2]$ metastable state of argon and krypton are experimentally determined using a magneto-optical trap. The method to determine the lifetime of a metastable state from a fluorescence decay measurement is described. The obtained lifetimes are 38^{+8}_{-5} s for argon and 39^{+5}_{-4} s for krypton.

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In this Letter we report the first measurement of a metastable state lifetime using a magneto-optical trap. A lifetime measurement requires that an atom be kept free from perturbation for an extended time. A standard method of measuring metastable lifetimes has been the time-of-flight (TOF) technique using an atomic beam [1–4]. Even for a helium beam, which could be operated at a very low temperature, it was found to be difficult to keep atoms free from perturbations beyond several tens of milliseconds [2]. The TOF measurement could give only a lower limit of 1 s for the lifetime of the $1s$ metastable rare-gas atoms [3]. As a result of the invention of atom traps one can now continuously observe atoms in free space for an extended time. Natural lifetimes of ions in metastable states exceeding a second have been measured using ion traps [5–10]. For neutral atoms, however, such measurement has not been reported to date. In the case of neutral atoms a difficulty arises, since the trapping potential directly modifies the internal state of trapped atoms.

Neutral rare-gas atoms can be trapped using a closed transition between the lowest excited metastable state $1s_5[np^5(n+1)s, {}^3P_2]$ and the higher $2p_9[np^5(n+1)p, {}^3D_3]$ state [11,12]. The decay rate of atoms in the trap can be easily determined from a fluorescence measurement of the $1s_5$-$2p_9$ transition. The fluorescence decay rate is not, however, equal to the natural decay rate of the $1s_5$ metastable state. Atoms in an optical trap are constantly illuminated by resonant light, and consequently stay partially in the upper state, $2p_9$, which has the transition rate to the ground state different from that of the metastable state, $1s_5$. Furthermore, collisions with residual gas molecules have a substantial effect on the measurement. Even at 10^{-10} Torr, the collision quenching rate is of the order of 10^{-2} s^{-1}, which is a non-negligible fraction of the natural decay rate of the $1s_5$ state.

We have used a two-step procedure to deduce the natural decay rate of the $1s_5$ metastable state from a fluorescence decay measurement. First, we modulated the trapping laser beam by a square wave at a frequency of 500 kHz to separate the decay of the $1s_5$ state from that of the $2p_9$ state. Since the $2p_9$ state decays to the $1s_5$ state in approximately 30 ns, the atom is in the $1s_5$ state while the laser is turned off. By repeating the measurement at various on-off duty ratios of the laser, and by extrapolating the results to zero duty ratio, we deduced the decay rate characteristic of the $1s_5$ state. We repeated the above process at various background pressures to eliminate the effect of collisions.

The actual experimental setup is shown in Fig. 1. A metastable atomic beam was generated by a glow discharge and was slowed in a Zeeman-tuned slowing section [13]. Atoms were tapped in a magneto-optical trap [14]. The optical configuration of the cooling and trapping laser beams was the same as that of Ref. [11]. A titanium-sapphire ring laser was used for the cooling and trapping. The laser beam was spatially filtered through a single-mode optical fiber and was divided into three trapping beams, each of which had a power density of 5 mW/cm^2 with a diameter of 10 mm and was tuned approximately 1.5 linewidth below the resonance. They were carefully aligned to form a nearly spherical cloud of atoms. We found, however, that the optical alignments and the cloud shape were not critical to determine the linear decay rate. This was not unexpected, since the depth of the trap obtained by the push-and-recapture technique [14] was approximately 1 K while the kinetic energy of the atoms was less than 1 mK. The number of trapped atoms was approximately 10^5 with a $1/e$ radius of 0.5 mm. A magnetic field gradient at the center of the

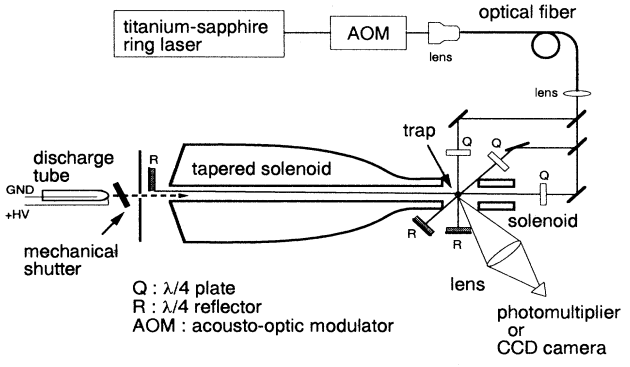

FIG. 1. Experimental configuration.

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trap was typically 1 mT/cm along the atomic beam axis. A mechanical shutter was placed immediately after the atomic source. The fluorescence from the trapped atoms was detected by a photomultiplier and processed by a photon counter. When the pattern of the trapped atoms was monitored, the photomultiplier was replaced by a charge-coupled device camera.

The experimental procedure to obtain the $1s_5$ natural lifetime was as follows. First, atoms were loaded into the magneto-optical trap. The atomic beam source was then shut off by the mechanical shutter, simultaneously with the start of the laser modulation at a particular duty ratio. After waiting for several seconds to stabilize the trap to the modulated laser, we started to measure the fluorescence intensity from the trapped atoms.

The decay of the fluorescence intensity I was fitted to the solution of the equation describing a sum of linear and quadratic decaying terms,

$$dI/dt = -\alpha I - \beta I^2 . \quad (1)$$

The mean relative deviation between the experimental points and the theoretical curve was less than 4% in the intensity range of 2.5 decades that was used for the curve fitting. The relative fitting error of an individual experimental run was less than 10% for α and β.

The quadratic term in Eq. (1) arises from collisions between trapped atoms. The linear term is a sum of the natural decay rate γ of the atom in the mixed state $1s_5$-$2p_9$ and the decay rate kn_p resulting from collisions with the residual gas.

$$\alpha = \gamma + kn_p , \quad (2)$$

where k is the collision rate constant and n_p is the density of background gas molecules at pressure p.

We measured the fluorescence decay of ^{40}Ar, ^{84}Kr, and ^{87}Rb using the same experimental setup. Rubidium was used to confirm the stability of the trap and to reduce the ambiguity of the metastable-state decay rate as discussed below. For this atom the trapping transition was $^2S_{1/2}(F=2)$-$^2P_{3/2}(F=3)$. A diode laser resonant to the $^2S_{1/2}(F=1)$-$^2P_{3/2}(F=2)$ was used to recycle the population that had decayed to the $F=1$ ground hyperfine state.

We repeated the same measurement at different duty ratios δ ranging from 0.125 to 1.0 to obtain the δ dependence of the linear decay rate α. Figure 2 shows the results for Ar, Kr, and Rb at a pressure of 2×10^{-10} Torr. For Ar and Kr, the value of α changed linearly with δ between $\delta=0.2$ and 1, and the diameter of the cloud of the trapped atoms was almost constant.

The two terms in Eq. (2) have different δ dependence. The mixing ratio of the $2p_9$ state is a linear function of δ. Therefore, the natural decay rate γ changes linearly with δ, and the value at $\delta=0$ is the natural decay rate $\gamma^{(0)}$ of the $1s_5$ state. The trap potential depth decreases to zero when δ goes to zero, and the collision rate k will diverge at $\delta=0$. However, if the δ dependence of α is approximated by a linear function, the contribution of the second term in the equation for α will remain finite. We write this contribution as $k^{(0)}n_p$. The collision rate $k^{(0)}$ is independent of n_p if the fitting range is fixed for all measurements. We define the linear decay rate $\alpha^{(0)}$ as the value of α that is obtained by a linear extrapolation to $\delta=0$ from the measurements between $\delta=0.2$ and 1.0. In the following descriptions, the superscript indicates the duty ratio δ and the subscript the pressure p of foreign gas introduced into the trap.

We repeated the entire experimental procedure to obtain $\alpha^{(0)}$ at various gas pressures in the trap. The collision rate constant $k^{(0)}$ was determined from the slope of the pressure dependence of $\alpha^{(0)}$. If the composition of the residual gas does not change, the value of $\alpha^{(0)}$ at $p=0$, $\alpha_0^{(0)}$ should be equal to the natural decay rate of the $1s_5$ metastable state. However, the gas composition at the lowest pressure was unknown, and a careful analysis of the data was necessary. The mass spectrum of the residual gas showed that it was mostly composed of hydrogen, hydrocarbons, CO, or N_2. We used a liquid nitrogen trap during the operation of the experiment so that water and CO_2 were not significant near the trap. We measured the pressure dependence using H_2, CH_4, N_2, Ar, and Kr as foreign gases and determined $k^{(0)}$ for Ar

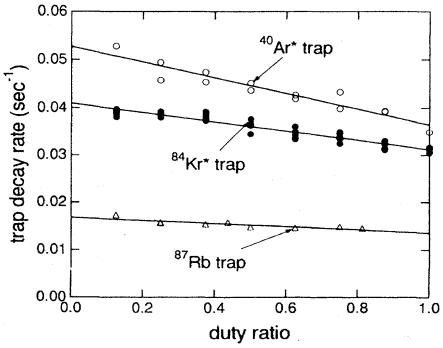

FIG. 2. The duty ratio δ dependence of the trap decay rate at the pressure 2×10^{-10} Torr.

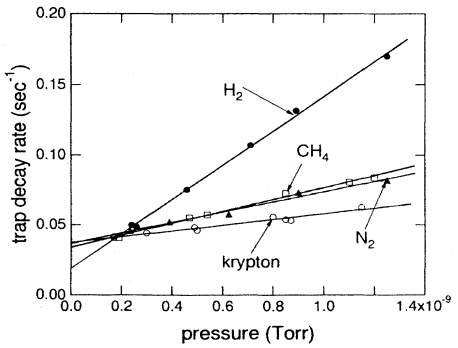

FIG. 3. The pressure dependence of the decay rate $\alpha^{(0)}$ for Kr.

and Kr. An example of the pressure dependence of $\alpha^{(0)}$ is shown in Fig. 3. Table I summarizes the results for $k^{(0)}$. In Fig. 2 and Table I the pressure is the nominal value obtained from the ionization gauge (ANELVA NIG-2F). Since H_2 has the lowest sensitivity on the gauge among possible residual gas species [15] the natural decay rate $\gamma^{(0)}$ should be in the range between $\alpha_0^{(0)}$ obtained by introducing H_2 as a foreign gas and $\alpha^{(0)}$ at the attainable minimum pressure (2×10^{-10} Torr). The values obtained by this procedure are between 0.0179 and 0.0419 s^{-1} for Kr, and between 0.0152 and 0.0512 s^{-1} for Ar.

The error range of the above results can be narrowed from a more careful consideration of the effect of the residual gas with the help of the Rb measurement. Table I also lists the collision rate constants for Rb at $\delta=0.125, k^{(0.125)}$. Since the natural lifetime of Rb is infinite, the α_0 should scatter around the origin. To obtain α_0 for Rb, we measured $\alpha_p^{(\delta)}$ at various duty ratio δ and pressure p, and extrapolated the values to $p=0$ for each δ. The extrapolated lines for different δ intersected over a small range of decay rates whose values depended only on the foreign gas species. We obtained $\alpha_0(\mathrm{Rb{:}N_2})=(9.1\pm1.2)\times10^{-3}$ s^{-1} and $\alpha_0(\mathrm{Rb{:}H_2})=-(8.4\pm1.5)\times10^{-3}$ s^{-1}, where the first abbreviation in parentheses indicates the atom in the trap and the second the foreign gas introduced to extrapolate α to zero pressure. The uncertainties are primarily due to the scattering of the extrapolated value on δ. This result together with the trap stability of α against the optical alignment and the duty ratio δ convinced us that the trap was operating as expected and did not have an intrinsic decay.

TABLE I. The collision rate of the trapped atoms with foreign gases, $k^{(0)}$ for Ar and Kr, and $k^{(0.125)}$ for Rb. The pressure is the reading of the ionization gauge.

Foreign gases	Collision rates (10^7 Torr^{-1} s^{-1}) ^{40}Ar*[$k^{(0)}$]	^{84}Kr*[$k^{(0)}$]	^{87}Rb[$k^{(0.125)}$]
Kr	...	2.1	...
Ar	3.3	...	...
N_2	6.3	3.7	4.5
CH_4	4.8	4.3	...
H_2	18	12	15

Table I shows that the ratios of the collision rate k for various foreign gases do not have a large difference for Kr and Rb. This is expected since the trapped atoms are kicked out of the trap by collisions with a large impact parameter. In such a case the collisions are dominantly elastic, and their cross section depends mostly on the configuration of the outermost electron. Our result is also justified from the state unresolved measurement by Ref. [16]. Therefore, the natural decay rate $\gamma^{(0)}(X)$ of Kr can be determined from the interpolation between $\alpha_0^{(0)}(X{:}N_2)$ and $\alpha_0^{(0)}(X{:}H_2)$ using the same weighting factor that produces $\gamma(X)=0$ for Rb, i.e.,

$$\gamma^{(0)}(X)=\frac{\alpha_0(\mathrm{Rb{:}N_2})\alpha_0^{(0)}(X{:}H_2)-\alpha_0(\mathrm{Rb{:}H_2})\alpha_0^{(0)}(X{:}N_2)}{\alpha_0(\mathrm{Rb{:}N_2})-\alpha_0(\mathrm{Rb{:}H_2})}, \tag{3}$$

where X means the species of the trapped atoms.

Another possible source of error is the quenching of the metastable state by a magnetic field. A rough estimate of the mixing ratio with fast decaying states by a trapping magnetic field shows that the decay due to the mixing is less than 1% of the natural decay rate. We carried out the decay measurement by varying the magnetic field by a factor of 4. A difference beyond the fitting error of 10% was not observed, although a slight increase with the magnetic field was found.

The natural decay rate for Kr obtained from Eq. (3) is $\gamma^{(0)}(\mathrm{Kr})=0.0258\pm0.0029$ s^{-1}. The mean value agrees with the difference of the decay rate γ at the minimum pressure between Kr and Rb in Fig. 2. This shows that the contribution of residual gas collisions on α is approximately the same for Kr and Rb. To obtain $\gamma^{(0)}$ for Ar, we use the same equation (3) as for Kr. This is justifiable since the ratio of the collision rates among various gases is not far from proportional and that the error arising from the ambiguity of the gas composition is estimated within other sources of errors. The value for Ar is then $\gamma^{(0)}(\mathrm{Ar})=0.0264\pm0.0044$ s^{-1}. The largest source of the error was the fitting error of α and the deviation of $k^{(0)}$ from the proportionality law among Rb, Ar, and Kr.

Our results are 32% and 56% for Ar and Kr, respectively, smaller than the theoretical values of Small-Wallen and Chou Chiu [17]. Their values, however, are based on the theoretical decay rate of the $1s_2$ state by Dow and Knox [18]. If the Small-Wallen value for Ar is corrected using the experimentally determined value for the $1s_2$ state of 5×10^8 s^{-1} by Lawrence [19], the discrepancy between theory and the present experiment is reduced to less than 12%.

In conclusion, we have demonstrated natural lifetime measurements of metastable states that are much longer than 1 s using the recently developed optical trap of neutral atoms. Although atoms in the trap are constantly interacting with a strong resonant light, we have shown that it is possible to deduce the natural decay rate of a specific state of the atom with a reasonably high accuracy. The same technique can be applied to metastable states that can be trapped by optical or electromagnetic means. Examples are the $1s_5$ state of other rare gases, the $^3P_2$ and $^3D_2$ states of alkaline earth atoms, and the $^3P_1$ state of the ground state multiplet of group-IV atoms. Our experimental procedure also provides infor-

mation on the collision characteristics of the metastable atom. If the density and kinetic energy of the trapped atoms and the depth of the trap potential are calibrated, the energy-resolved collision rate at room temperature as well as at extremely low temperatures can be determined from our measurement. A detailed description of the collision characteristics of metastable Ar and Kr will be given elsewhere.

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