Lifetime Measurement of the $^3P_2$ Metastable State of Strontium Atoms

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We have measured the lifetime of the $5s5p\,^3P_2$ metastable state of strontium atoms by magneto-optically trapping the decayed atoms to the ground state, which allowed sensitive detection of the rare decay events. We found that the blackbody radiation-induced decay was the dominant decay channel for the state at $T = 300$ K. The lifetime was determined to be 520^{+310}_{-140} s in the limit of zero temperature, arguably the longest lifetime ever determined in a laboratory environment.

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Precision measurements require the isolation of the physical system under study from environmental perturbations. The long coherence time thus obtained can be exploited to carry out ultrahigh resolution spectroscopy [1] or to create and control the macroscopic quantum coherence in atomic systems, such as Bose-Einstein condensate (BEC) [2] and Cooper pairing [3]. Furthermore, the origin of environmental decoherence has recently attracted much attention in the context of quantum computation/communication, in which an entangled quantum system should maintain its internal and/or motional state coherence [4,5]. The lower-lying ^3P metastable states [6–12] of alkaline-earth species are intriguing candidates for these studies. A long metastable lifetime may allow an optical spectroscopy at the 1 mHz level [13], enabling one to realize an ultraprecise atomic clock. In addition, the possibilities of evaporatively cooling them to reach BEC are discussed theoretically [14,15].

Laser-cooled and trapped atoms in ultrahigh vacuum condition offer an ideal sample for these studies, as the collisional interactions with container walls or residual gases can be substantially removed. However, in some cases, the radiation from the surrounding walls dramatically affects the evolution of the internal state coherences. The room-temperature blackbody radiation (BBR) has its intensity peak around a wavelength of 10 μm. Such BBR manifests itself most in Rydberg states [16–18], where infrared transitions with large electric dipole moments can be found. However, because of large energy differences, it hardly excites atoms in the lower-lying states. Its influence, therefore, has been rarely discussed in laser cooling and trapping. On the other hand, it is known that the BBR causes small but non-negligible ac Stark shifts in ultraprecise spectroscopy [19,20].

As for the ^3P metastable state of heavier alkaline-earth atoms, the upper-lying ^3D states are connected to the metastable state by electric dipole transitions with mid-infrared wavelength. Therefore, the excitation of the metastable state by the BBR may significantly alter its effective lifetime or introduce detectable blackbody shifts in precision spectroscopy. In this Letter, we study the influence of the room-temperature BBR [21] on the

metastable state lifetime of Sr [22], which is recently predicted to be 1050 s [23]. Because the lifetime is significantly longer than the collision-limited lifetime of tens of seconds that is realized in neutral atom traps at a vacuum pressure of 10^{-10} torr [24], observing the survival of metastable atoms in such traps will not be practical [24,25]. Instead of directly observing the decay of the metastable atoms by emitted photons [26], we monitored the occurrence of the rare decay events to the ground state by magneto-optically trapping the atoms on the $^1S_0 - {}^1P_1$ transition. In this way, we used the magneto-optical trap (MOT) as a photon amplifier with a gain of 10^{10} to sensitively detect the decay events [27,28].

Figure 1 shows the relevant energy levels for ^{88}Sr. Three radiative decay channels from the metastable $5s5p\,^3P_2$ state have been identified by Derevianko [23]: (1) The $M2$ transition to the ground state, (2) decay to the $^3P_1$ state through the $M1$, $E2$, and $M3$ transition, (3) decay to the $^3P_0$ state through the $E2$ transition, where En or Mn stands for an electric or a magnetic 2^n-pole transition. The theory suggests that the contribution of the channel (3) is only 0.1% [23]. Therefore, 99.9% of the population in the $^3P_2$ state finally relaxes to the $^1S_0$ ground state, as the atoms that decayed to the $^3P_1$ state further decay to the $^1S_0$ ground state in 22 μs. On this basis, we use the $^1S_0$ state population to detect the decay of the $^3P_2$ state.

We measured the number of atoms $N_S(t)$ in the $^1S_0$ ground state by applying the MOT on the $^1S_0 - {}^1P_1$ transition. The change of the atom number in the MOT is given by the rate equation,

$$\frac{d}{dt}N_S(t) = -(\gamma_p + \Gamma_c)N_S(t) + (\gamma_r + \Gamma_q)N_P(t). \quad (1)$$

The atoms in the ground state are collisionally lost at a rate $\Gamma_c \approx (10 \text{ s})^{-1}$ or leaked out of the MOT transition via the $5s4d\,^1D_2$ state with the typical rate of $\gamma_p \approx (14 \text{ ms})^{-1}$. The atoms are supplied from the magnetically trapped $^3P_2$ metastable state with its atom number of $N_P(t)$, which either radiatively decay (γ_r) or collisionally quench (Γ_q) to the ground state. Although the dominant
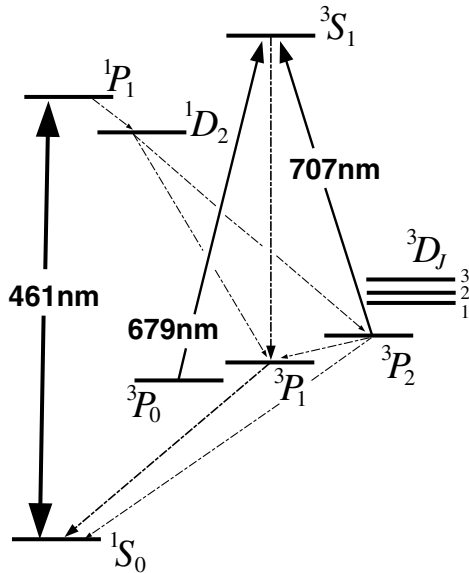
FIG. 1. The relevant energy levels for ^{88}Sr. The $^1S_0 - {^1P_1}$ transition at $\lambda = 461$ nm is used for trapping and detection. The leak path through the $^1D_2$ state is used for loading atoms into the $^3P_2$ metastable state. The $^3P_2 \rightarrow {^3S_1}$ transition at $\lambda = 707$ nm and the $^3P_0 \rightarrow {^3S_1}$ transition at $\lambda = 679$ nm are used to transfer the $^3P_2$ state population into the ground state. The upper-lying ^3D states are connected to the ^3P states by the electric dipole transitions at $\lambda \approx 3 \, \mu$m.

decay channel for $N_P(t)$ is the collisional ejection from the magnetic trap, this rate $\Gamma_m \approx (10 \text{ s})^{-1}$ is nearly 1000 times smaller than γ_p that governs the dynamics of $N_S(t)$. Therefore, the population of the ground state adiabatically follows that of the metastable state. In the time scale of interest, i.e., $t \gg \gamma_p^{-1}$, the ground state population is given by

$$N_S(t) = N_P(t) \frac{\gamma_r + \Gamma_q}{\gamma_p + \Gamma_c}. \qquad (2)$$

In the following, assuming that $\Gamma_c / \gamma_p \ll 1$ and $\Gamma_q / \gamma_r \ll 1$ hold as discussed later, we neglect both of the collisional loss terms in Eq. (2). The metastable decay rate γ_r, therefore, can be determined by the ratio $N_S(t)/N_P(t)$ of population distributed in both of the states and the MOT decay rate γ_p. The number of atoms in the $^1S_0$ ground state can be derived by observing the MOT fluorescence intensity of $I_S(t) = \eta N_S(t)$, where η is the photon counting rate per atom trapped in the MOT. Similarly, $N_P(t)$ can be obtained by transferring the metastable state population into the ground state and measuring the MOT fluorescence intensity of $I_P(t) = \eta N_P(t)$. By the ratio of these fluorescence intensities, $\gamma_r / \gamma_p [= I_S(t)/I_P(t)]$ is accurately determined regardless of the coefficient η.

The apparatus for magneto-optically trapping strontium atoms is similar to that described in Ref. [29]. The MOT fluorescence was collected by a lens with a solid angle of 10^{-5} and then sent to a photomultiplier tube (PMT). An interference filter was placed in front of the PMT to block except the 461 nm light. The output signal

was then sent to a multichannel scaler. The two transitions, $^3P_2 \rightarrow {^3S_1}$ and $^3P_0 \rightarrow {^3S_1}$ (Fig. 1), were used to pump the $^3P_2$ state population into the $^1S_0$ ground state.

We first magneto-optically trapped atoms on the $^1S_0 - {^1P_1}$ transition. In this trapping period of 0.3 s, the $^3P_2$ metastable state was populated via the weak branching decay to the $^1D_2$ state from the $^1P_1$ state, which is estimated to be 10^{-5} [30]. About 10^7 atoms in the low-field-seeking state were thus accumulated in the magnetic trap, which is formed by the quadrupole magnetic field used for the MOT. The field gradient was 100 G/cm along its axis of symmetry. At the typical peak density of $\approx 10^9 \text{cm}^{-3}$, two-body collisional loss rate is estimated to be much smaller than the collisional loss term Γ_c in Eq. (1), where we assumed an inelastic collisional loss rate of $\approx 10^{-11} \text{cm}^3/\text{s}$ [14,15]. We then turned off the MOT lasers and closed the shutter that blocked both the atomic beam and the thermal radiation from the oven. We waited for 0.3 s so that all atoms except magnetically trapped metastable atoms diffused out of the trap region. After that, we turned on the MOT lasers again to capture atoms that were radiatively decayed from the $^3P_2$ state and recorded the fluorescence intensity. At $t = 1$ s, we irradiated both of the pumping lasers to transfer the metastable state population into the $^1S_0$ ground state and determined the number of atoms trapped in the $^3P_2$ state by the MOT fluorescence intensity.

Figure 2 shows a change of fluorescence intensity averaged over 10^2 measurements, where the background level was subtracted by alternating the same procedure with and without loading atoms into the magnetic trap. The number of atoms in the $^1S_0$ state at $t = 1$ s was determined by exponentially extrapolating the whole decay curve $I_S(t)$ of the MOT fluorescence to obtain $I_S(1)$ with
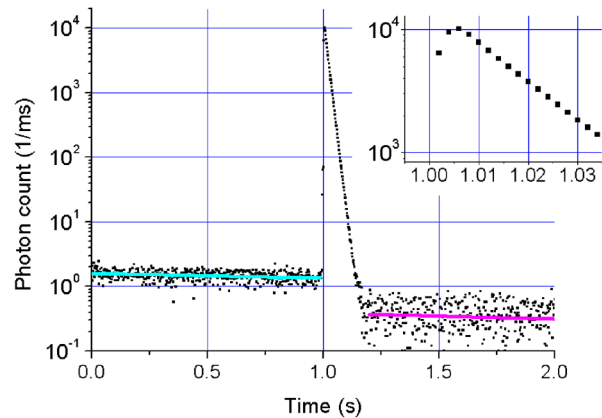

FIG. 2 (color online). The photon counting signal. The signal at $0 < t < 1$ s indicates the photons scattered by the recaptured atoms in the ground state. At $t = 1$ s, the $^3P_2$ metastable state population is pumped into the ground state. The peak intensity gives the number of atoms in the metastable state. The decaying slope gives the MOT lifetime, which is $\gamma_p^{-1} = 14.1$ ms. The inset shows the enlarged figure around the peak. The fluorescence intensity reaches its maximum in ≈ 5 ms.

better statistics. The fluorescence decay in $0 < t < 1$ s was mainly caused by the collisional atom loss in the magnetic trap with the decay rate $\Gamma_m = (6.6 \text{ s})^{-1}$ at the background gas pressure of 6.6×10^{-10} torr. By transferring the metastable state population into the ground state at $t = 1$ s, the fluorescence intensity sharply rose up to its maximum in about 5 ms. We approximated $I_P(1)$ by the peak fluorescence intensity shown in the inset of Fig. 2. The signal then decayed double exponentially, consisting of the MOT decay with $\gamma_p = (14.1 \text{ ms})^{-1}$ due to the branching loss and the much slower collisional decay Γ_m of the metastable atoms recaptured in the magnetic trap. The metastable state lifetime is calculated by applying the ratio $N_S(1)/N_P(1) = I_S(1)/I_P(1)$ and the measured MOT decay rate γ_p in Eq. (2). The measurement shown in Fig. 2 gave an effective radiative lifetime of $\gamma_r^{-1} = 104^{+8}_{-7}$ s (errors indicate purely statistical 1 standard deviation), which is only one tenth of the theoretical lifetime [23]. This shortening can be attributed to the BBR-induced decay via the $5s4d\,^3D$ state as discussed later. However, before discussing the BBR-induced decay, we checked the other decay channels.

The reduction of the lifetime may be caused by the fine structure mixing of the metastable $^3P_2$ state with the $^3P_1$ or the $^1P_1$ state in the presence of the trapping magnetic field. Assuming a magnetic field of 10 G, the magnetically induced decay rates are estimated to be 0.1% of the natural decay rate γ_0 of the $^3P_2$ state. Actually, we measured the metastable lifetime under various magnetic field gradient, however, no magnetic-field-dependent change was found. We note that there are no hyperfine structure induced decay channels, as the ^{88}Sr isotope has no nuclear spin. Second, due to collisions with background gases, atoms in the metastable state may be (1) kicked out of the magnetic trap with the rate Γ_m or (2) quenched into the ground state with Γ_q. For the former issue, since we compared the fluorescence intensity I_S and I_P just before and after the population transfer, the collisional atom loss in the transferring period of $\tau_p = 5$ ms may cause an error. However, its fraction is estimated to be as small as $1 - e^{-\Gamma_m \tau_p} \approx 10^{-3}$. For the latter issue, the collisional quench to the ground state may cause pressure-dependent shortening of the metastable state lifetime. To check this influence on the measured lifetime, we increased the background gas pressure up to 1.6×10^{-9} torr. However, no pressure dependence was found, indicating that the contribution of Γ_q is well within 5% of the statistical uncertainty for γ_r.

The rapid decay can be attributed to the metastable state quenching due to the BBR field that transfers atom population in the $^3P_2$ state to the short-lived $^3P_1$ state via the ^3D states. By solving the coupled rate equations, the steady-state value of the BBR-induced decay rate $\gamma_B(T)$ is expressed as

$$\gamma_B(T) = \gamma_D \frac{7}{36} \bar{n}(T), \quad (3)$$

where γ_D is the radiative decay rate of the $5s4d\,^3D$ state, and $\bar{n}(T) = \{\exp(hc/k_B T \lambda) - 1\}^{-1}$ is the BBR photon occupation number at temperature T with the wavelength $\lambda \approx 3.01$ μm for the $^3P_2 - ^3D$ transition. Using the radiative lifetime $\gamma_D^{-1} = 2.9 \pm 0.2$ μs of the ^3D states [31], the BBR-induced decay rate is calculated to be $\gamma_B(T_0) = 8.03 \times 10^{-3}$ s^{-1} for $T_0 = 299.5$ K. This is nearly an order of magnitude larger than the theoretical decay rate, therefore can explain the observed lifetime shortening. To confirm that this lifetime shortening originates from the BBR excitation, we measured the decay rate $\gamma_r(T)$ as a function of the ambient temperature T by heating up the vacuum chamber that enclosed the MOT. We note that this temperature change altered the vacuum pressure in the range of $(6.6–9.7) \times 10^{-10}$ torr. However, the resultant collisional losses did not affect the measured lifetime as mentioned previously. The measured decay rate $\gamma_r(T)$ is plotted in Fig. 3 by filled squares, where error bars indicate 1 standard deviation. The monotonic increase of the decay rate as the temperature clearly supports that this lifetime shortening is caused by the BBR.

The BBR-induced decay rate is estimated as follows: Initially, the chamber that surrounds the atoms is assumed to be in thermal equilibrium. However, in heating up the chamber, care should be taken if it is in thermal equilibrium. The vacuum chamber is made of stainless steel with 150 mm diameter and has six viewing ports made of BK7 glass with 40 mm diameter. It was heated by a ribbon heater wound around the chamber body. We monitored the temperature of the viewing ports (at their rim and center) and the body of the chamber 1 h after changing the heating power: Whereas the temperature of the rim was the same as that of the chamber body T, that of the center was somewhat lower by $\delta T(> 0)$. We therefore defined an average temperature for the viewing ports as $T_p = T - \delta T/2$, where we assumed a quadratic variation of their temperature in the radial direction as
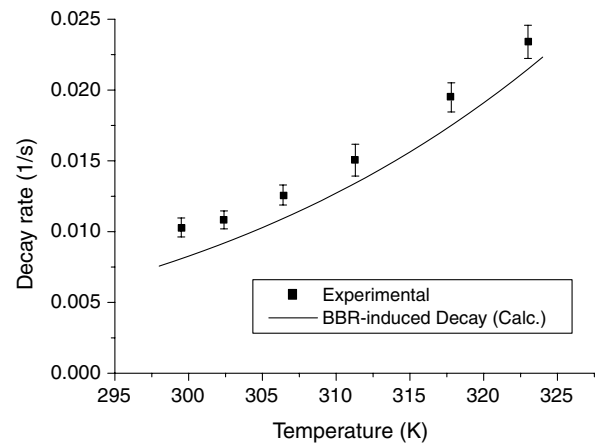

FIG. 3.   Measured decay rate γ_r under different ambient temperature T. The calculated BBR-induced decay rate $\gamma_B(T)$ is also shown. The offset between the measured data points and the calculated line indicates the natural decay rate γ_0.

confirmed by numerically solving the heat transfer equation using the finite element method. Owing to this temperature inhomogeneity, atoms are not in the BBR field of thermal equilibrium. Assuming a spherical radiant cavity, the effective photon occupation number for the body temperature of T is calculated as [32]

$$\bar{n}_{\text{eff}}(T) = \frac{\Omega_b \varepsilon_b \bar{n}(T) + \Omega_p \varepsilon_p \bar{n}(T_p)}{\Omega_b \varepsilon_b + \Omega_p \varepsilon_p}. \tag{4}$$

Here Ω_b is the solid angle covered by the chamber body and ε_b is its spectral emissivity at $\lambda \approx 3~\mu$m. Likewise, we defined Ω_p and ε_p for the viewing ports, which fraction in the solid angle was $\Omega_p/\Omega_b \approx 10^{-1}$. We used this effective photon occupation number $\bar{n}_{\text{eff}}(T)$ to calculate the BBR-induced decay rate given by Eq. (3). We took the spectral emissivity of both objects as fitting parameters so that the measured decay rate $\gamma_r(T)$ should have a constant offset γ_0 to the calculated BBR-induced decay rate $\gamma_B(T)$ at each temperature. After the least squares fitting, we obtain the metastable decay rate of $\gamma_0 = (1.9 \pm 0.7) \times 10^{-3}~\text{s}^{-1}$. Here the statistical error of $\pm 0.4 \times 10^{-3}~\text{s}^{-1}$ was given by the fitting with spectral emissivities $\varepsilon_b = 0.3\%$ and $\varepsilon_p = 92\%$ for stainless steel and BK7 glass, respectively [33], whereas the larger uncertainty of $\pm 0.6 \times 10^{-3}~\text{s}^{-1}$ was introduced from the radiative lifetime of the ^3D state [31] in correcting $\gamma_B(T)$.

In summary, by carefully subtracting the influence of the blackbody radiation, we have determined the natural lifetime of the $^3P_2$ state of ^{88}Sr to be 520^{+310}_{-140} s, which showed fair agreement with the theoretical value of 1050 s [23]. The deduced lifetime, which is arguably the longest one determined from a state decay measurement in a laboratory [34], is comparable to that measured in nebular spectra [35], indicating that effectively high reduction of perturbation can be done for trapped atoms. The influence of the room-temperature BBR on the metastable state lifetime has often been overlooked so far, as the time scale was far beyond reach for experiments [34]. However, these previously inaccessible time scales are becoming practically important in recent experiments of forming a BEC [14,15] or ultrastable atom clocks [13,34]. To fully access the natural lifetime in the present case, cold environment needs to be prepared: e.g., a stringent comparison of the $^3P_2$ state lifetime with the theory [23] can be possible by lowering the ambient temperature down to 217 K, where the BBR quenching rate is expected to be 1% of the natural decay rate.

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