Direct measurement of the metastable $^3P_2$ decay rate in krypton

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We use a magneto-optical trap to isolate ^{84}Kr atoms in the metastable $^3P_2$ state and then measure the rate of 125.0-nm fluorescence due to spontaneous magnetic quadrupole ($M2$) decay. We normalize the overall fluorescence rate from the sample using a ''quenching'' process in which laser excitation provides a path for prompt decay to the ground state, with each atom emitting one 123.6-nm photon. A single, solar-blind photomultiplier tube is used to detect both the $M2$ and the quench-induced photons, so that only small corrections relating to spectral response are necessary. We find a spontaneous decay rate of $\Gamma = 0.0354(22)\ s^{-1}$, which translates into a lifetime of 28.3(1.8) s. This result is discussed in the context of previous measurements and calculations of $^3P_2$ lifetimes in the rare gases.

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INTRODUCTION

The lowest electronic excited state in any rare-gas atom heavier than He is well described as a $^3P_2$ state in *L-S* coupling. In the absence of hyperfine structure, its spontaneous decay to the $^1S_0$ ground state is allowed, at lowest order, through the magnetic quadrupole ($M2$) interaction. The state is therefore metastable, with an expected lifetime of many seconds. Theoretical calculations disagree considerably as to the actual lifetime values, but they consistently generate values that increase as one descends the periodic table of elements [1,2]. For instance, assuming a particular form for electron correlations in a relativistic model, Indelicato *et al.* calculate $^3P_2$ lifetimes of 22, 51, 63, and 96 s for Ne, Ar, Kr, and Xe, respectively.

Accurate lifetime measurements on such long time scales have been possible with neutral atoms only since the introduction of laser-cooling and -trapping techniques, which provide the necessary state and isotope selectivity as well as long observation times. Katori and Shimizu first pursued such measurements with magneto-optically trapped Ar and Kr [3]. Their strategy was to monitor the loss of atoms from a modulated magneto-optical trap (MOT) and extract that portion of the loss rate contributed by the $^3P_2$ decay. This method was complicated by the need for extrapolation to a MOT-on duty cycle of zero and to a background-gas pressure of zero, but it nevertheless yielded lifetimes of 38 s for ^{40}Ar and 39 s for ^{84}Kr, with uncertainties of 10–20% quoted in both cases.

Walhout *et al.* developed an alternative, photon-counting technique in order measure metastable lifetimes in Xe [4]. They measured the rate of vacuum-ultraviolet (VUV) fluorescence from the $^3P_2$ decay of Xe atoms in a MOT. The rate was indexed to the number of trapped atoms by a $^3P_2$-quenching process that forced every trapped atom to emit a single VUV photon. A single photomultiplier tube was used to detect photons from both the $M2$ decay and the quench process. Apart from small corrections related to the angular distribution of photons and the detector's spectral response, the $^3P_2$ decay rate was simply obtained from the ratio of the total $M2$ decay rate to the number of detected quench-induced photons. The lifetime for ^{132}Xe, or the inverse of the measured decay rate, was determined to be 42.9(0.9) s.

The experimental lifetimes for Ar, Kr, and Xe generally disagree with theoretical calculations. The disparity is seen not only in the absolute lifetime values, but also in the dependence of the $^3P_2$ lifetime on the atomic number (Z). The nonrelativistic $M2$ transition probability scales as the fifth power of the transition energy [5], and this energy decreases significantly with increasing Z. The Z dependence of the lifetime is therefore expected to be quite pronounced, even after relativistic corrections are applied. However, the trend in the measurements just cited does not match this expectation, and the situation has both experimentalists and theorists double-checking their results.

We report here a measurement of the $^3P_2$ decay rate in ^{84}Kr. We use the photon-counting method of Ref. [4] and find a lifetime significantly shorter than that found with the trap-decay method of Ref. [3]. When combined with the earlier Xe measurement, our result confirms the theoretical prediction of clear Z dependence in the rare-gas $^3P_2$ lifetimes. However, it does not erase long-standing concerns over the apparently inflated numerical estimates of the absolute lifetime values.

EXPERIMENTAL SETUP

Our experiments start with an atomic Kr beam emerging from a boron nitride nozzle that is cooled to 77 K. A dc discharge running through the nozzle excites some of the atoms into the $^3P_2$ state, and these undergo Zeeman-tuned laser cooling and deceleration [6,7] before being trapped in a standard, six-beam MOT [8]. A 40-mW master diode laser is grating stabilized [9] and locked to the laser-cooling resonance at 811.3 nm. Its light is divided between several acousto-optic modulators, which generate beams with tun-

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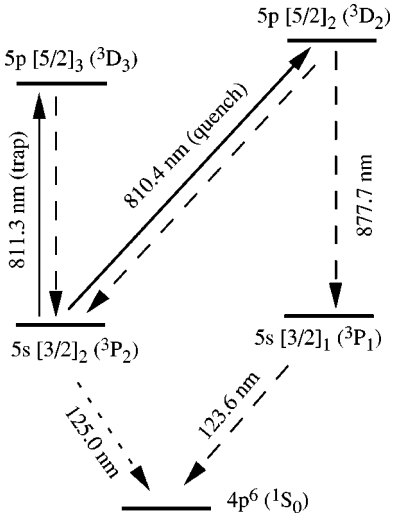

FIG. 1. Transitions used in the measurement of the $^3P_2$ decay rate in ^{84}Kr. Solid lines represent laser excitations, dashed lines are electric-dipole decays, and the dotted line corresponds to the weak M2 decay. States are specified in pair-coupling scheme, in which the core angular momentum (J_c) is coupled to the orbital angular momentum (l) of the valence electron to give $K=J_c+l$, and K is coupled to the valence spin (s) to produce a total angular momentum J. The designation in this scheme is $nl[K]_J$ or $nl'[K]_J$, where l is unprimed for $J_c=3/2$ and primed for $J_c=1/2$. Approximate L-S-coupling designations are indicated in parentheses.

able frequency offsets. Two of the frequency-shifted beams are used for injection-locking a pair of additional diode lasers, one supplying the Zeeman-slowing beam and the other the trapping beams. The slowing beam is tuned below resonance by 300 MHz (56 linewidths) and directed through the trapping region. It is resonant with the decelerating atoms as they traverse the 40-cm-long slowing region, where the tuning field varies from zero near the Kr source to a maximum of 21.5 mT. The MOT is located 20 cm downstream from this field maximum. The 2-cm-diameter MOT beams are detuned by $\sim$10 MHz, and each typically has an intensity of 8 mW/cm^2.

A fourth diode laser is tuned to the 810.4-nm ''quenching transition'' and, by activation of a shutter, can be used to deexcite atoms from the $^3P_2$ state, forcing each atom to emit a single 123.6-nm photon as it decays through the short-lived $^3P_1$ state (see Fig. 1). We use a polarizing beam splitter to combine the quenching laser with the trapping laser, so that we can divide, steer, and retroreflect the two with the same optical elements. The quenching light is thus applied along the six directions of the MOT beams, but with opposite circular polarizations. The polarization of either laser field varies on spatial scales shorter than 800 nm—a fact that helps erase any net atomic orientation or alignment and therefore any anisotropy in the overall distribution of fluorescence. Moreover, if the quenching light optically pumps an atom into a nonabsorbing state, the scrambled polarization ensures that the atom will be recoupled to the laser after it moves a short distance.

From measurements of the quench-induced VUV signal, we estimate the typical number of trapped atoms to be nearly 10^6. By adjusting the MOT's field gradient between 0.1 and 1.5 mT/cm, we can vary the volume of the trapped cloud by a factor of about 300 and simultaneously change the atomic density by a factor between 10 and 30. When loading is interrupted, the MOT decays with a characteristic time of approximately 1 s because of collisions with background gas, the pressure of which is normally about 3×10^{-8} Torr.

Our goal is to monitor the total rate of 125.0-nm $M2$ fluorescence from atoms in the $^3P_2$ state and then to normalize that rate (that is, to index it to the number of trapped atoms) using the quench-induced fluorescence. Both the $M2$ and the quench-induced VUV signals pass through the same MgF_2 window and are detected with a single, solar-blind photomultiplier tube (PMT). We purge the 4-mm gap between the window and the PMT with argon gas in order to reduce atmospheric absorption. The PMT delivers single-photon pulses through a discriminator/amplifier to the input of a multichannel scalar (MCS). The ratio of the $M2$ and quench-induced signals is free of geometry-related factors, so the only corrections related to the detection scheme are small variations in PMT quantum efficiency and window transmission over the 1.4-nm range between the two detected wavelengths.

MEASUREMENT PROCEDURE

Our procedure calls for three measurement phases, in which we count photons from the $^3P_2$ decay, the quench process, and background. Data from all three of these phases are accumulated in a single 100-channel sweep of the MCS. The details of the measurement sequence are as follows.

To ensure that all atoms are in the $^3P_2$ state when we monitor the $M2$ decay, mechanical shutters are used to switch the MOT and slowing lasers at a 10-Hz frequency, with ''on'' and ''off'' intervals of 85 and 15 ms, respectively. The possibility of spurious signals from the atomic beam is eliminated by a shutter in the beam path, which we open for only 50 ms at the beginning of each ''on'' interval. Decay counts are accumulated only when the MCS gate is activated for 5–10 ms during each ''off'' interval. After each gated observation period, the MCS is advanced to its next channel and awaits the next gate signal.

When 50 of these observation cycles have been executed, the atomic beam shutter is closed, the quench laser is applied to the trapped cloud, and the resulting, prompt VUV photons are counted. The quench laser is detuned from resonance enough that the quenching process occurs over 20–30 ms, so pulse pile-up is not a problem. If, in the absence of the trapping beams, the quench duration is extended beyond 30 ms, the total VUV count decreases slightly, presumably because of ballistic loss. However, if the MOT beams are cycled as usual and therefore recapture atoms during the quenching process, this loss is not seen.

After each application of the quench laser, 50 MCS channels have recorded $M2$ decay counts, and five have recorded quench-induced counts. At this point, the MOT and/or slowing lasers are turned off, but the rest of the 10-Hz cycle resumes. During this time with no atoms in the trap, the VUV background rate is registered in the last 45 channels of

the MCS sweep. Normally, the lasers are then turned back on, and another sweep is initiated automatically, until the data from 20 MCS sweeps have been added together. The resulting data set leads to a single measurement of the decay rate, as described below.

ANALYSIS OF RESULTS

We designate the number of accumulated metastable-decay counts as N_M, the number of quench-induced counts as N_Q, and the number of background counts as N_B. N_M and N_B are each obtained over an elapsed time designated as Δt (typically the sum of 40 gated counting intervals). From these measurements we obtain a normalized count rate of

$$R=(N_M-N_B)/N_Q\Delta t.$$

This ''raw'' rate will differ from Γ by only about 10%, but several experimental details must still be considered. We will represent the total number of atoms addressed in the experiment by $\mathcal{N}$, the PMT quantum efficiency at wavelength λ (in nm) by $\mathcal{E}_\lambda$, the MgF_2 window transmission at wavelength λ by T_λ, and the detector solid angle by Ω. We will also account for some unwanted quenching of the $^3P_2$ state at rate R_{BB}, which may arise from magnetic-dipole excitation of the $^3P_1$ state by blackbody radiation. Finally, we will include a small correction for the $\sim$0.5(1)% of the atoms that are quenched through the $^1P_1$ and $^3P_0$ states and therefore go uncounted in the quench signal [10]. This correction requires that a correction factor of $\beta=0.995(1)$ be included in N_Q. The net (unnormalized) count rate for the metastable decay can now be written as

$$(N_M-N_B)/\Delta t=\mathcal{N}(\Omega/4\pi)[\Gamma\mathcal{E}_{125.0}T_{125.0}+R_{\mathrm{BB}}\mathcal{E}_{123.6}T_{123.6}],$$

and the total number of quench counts as

$$N_Q=\mathcal{N}\beta(\Omega/4\pi)\mathcal{E}_{123.6}T_{123.6}.$$

Solving for the decay rate Γ, we arrive at

$$\Gamma=\left\{\frac{\mathcal{E}_{123.6}T_{123.6}}{\mathcal{E}_{125.0}T_{125.0}}\right\}(\beta R-R_{\mathrm{BB}}).$$

From the manufacturer's calibration data for our Hamamatsu R6835 PMT, we estimate $(\mathcal{E}_{123.6}/\mathcal{E}_{125.0})=0.953(15)$. Similarly, calibration data for our Harshaw-Bicron MgF_2 window allow us to estimate $(T_{123.6}/T_{125.0})=0.971(9)$. Although the magnetic-dipole matrix element between the $^3P_1$ and $^3P_2$ states is not known with great accuracy, an adequate estimate of the blackbody rate at room temperature comes from standard perturbation theory [11], which suggests $R_{\mathrm{BB}}=0.0003(2)\ \mathrm{s}^{-1}$.

From more than 200 measurements of R, we find an average normalized count rate $R_{\mathrm{av}}=0.0388(21)\ \mathrm{s}^{-1}$. This result reflects photon-count accumulations of $\Sigma(N_M)\approx 34\,000$ and $\Sigma(N_B)\approx 4000$, each obtained over a net (gated) observation time of nearly 1600 s. The corresponding quench total of $\Sigma(N_Q)\approx 2\times 10^6$ is the result of $\sim$4000 applications of the quench laser. Some of the data included here are from runs in which possible systematic errors were being studied but proved to make little or no difference in the measured value of R. Among the variables shown to be unimportant by the systematic studies are the atom density in the MOT, the background gas pressure, the PMT voltage, the MOT field gradient, and variations in the ellipticity of the MOT and quench beams. Moreover, the electric field from a channel-electron multiplier that is 7 cm away from the trap has no detectable effect. Similarly, an active ion-gauge filament 30 cm away from the trap (but not in direct line of sight) does not change the value of R, although it does increase the number of background counts.

In addition to a statistical uncertainty of $<$1%, our uncertainty in R_{av} comes from two dominant systematic effects. First, we are unable to eliminate the possibility that intensity imbalances in the MOT or quenching beams may introduce some anisotropy in the VUV signal. This possibility is not considered in our analysis, so we have quantified the corresponding uncertainty by imposing imbalances and looking for a change in R. These tests indicate that R changes by no more than $\pm$5%, and we take this to be the level of uncertainty related to possible anisotropy.

The second systematic uncertainty entering into R_{av} arises from the possibility of inaccurate counting of quench-induced photons. If the quench laser is tuned too close to resonance, counts may be lost to pulse pile-up in the detector, whereas if it is detuned too far from resonance, the quench process may be extended beyond the gated observation interval. Our approach to this problem is to ensure against pulse pile-up by using a relatively large detuning and gating the MCS for tens of milliseconds. Nevertheless, sometimes a small but non-negligible fraction of the quench signal is detected very near the end of the gate interval, and this suggests that some of the signal may go undetected. From this observation, we estimate the uncertainty in N_Q to be 2%. Assuming uncorrelated errors, we come to a combined uncertainty in R_{av} of 5.5%.

With all the necessary measurements in hand, we arrive at our final determination for the $^3P_2$ decay rate in ^{84}Kr: $\Gamma=0.0354(22)\ \mathrm{s}^{-1}$, which translates into a lifetime of 28.3(1.8) s.

DISCUSSION

We suspect that the discrepancy between our result and the 39-s lifetime obtained previously may stem from unconsidered effects in the earlier work. In particular, the analysis in Ref. [3] assumed linear extrapolations of the fluorescence decay rate to a background-gas pressure and a MOT duty cycle of zero. These extrapolations seem not to be fully justified by the data presented, which indicate that the rate is linear only for MOT-on duty ratios $>$20% and that the pressure-dependent rates all converge at $\sim 0.04\ \mathrm{s}^{-1}$, regardless of background gas. Moreover, the analysis did not sufficiently differentiate between MOT-on and MOT-off conditions, which are known to involve different collisional loss constants [12,13]. Thus, it is conceivable that some systematic error may have found its way into the earlier result.

As suggested above, we have taken all reasonable steps

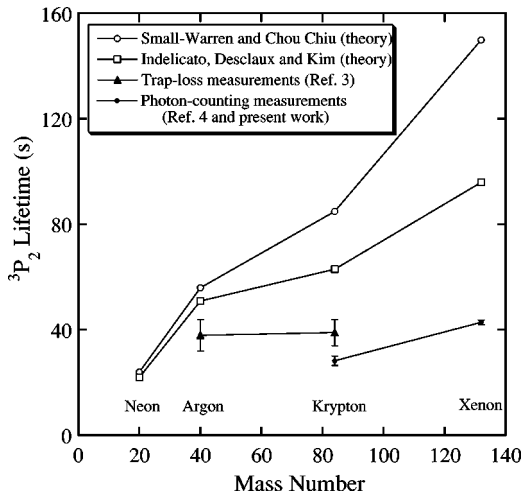

FIG. 2. Summary of experimental and theoretical $^3P_2$ lifetimes for the rare gases. Note that the photon-counting method used in this work and in Ref. [4] supports the theoretical expectation of clear Z dependence.

that we can think of in order to eliminate systematic biases in our result. We wish to emphasize that nonradiative collisions, which are of utmost concern in trap-loss measurements, cannot compromise our VUV-photon-counting technique. Of course, our analysis does not consider the possibility of nonexponential decay, a topic that has attracted considerable interest. In order for both Kr measurements to be correct, the $^3P_2$ state would have to decay faster at short time ($\sim$10 ms) than at long time ($\sim$10 s). A detailed discussion of this intriguing but seemingly unlikely scenario is beyond the scope of this paper.

Figure 2 gives an overview of some measured and calculated $^3P_2$ lifetimes for the rare gases. It is immediately obvious that the theoretical predictions lie significantly above the measured values, especially for the heavy elements. A leading explanation for this long-standing disagreement has been that the uncertainties in relativistic effects and electron correlations make it difficult to eliminate computational inaccuracies. These considerations are especially problematic in the heavier atoms. Very recently, Desclaux, Indelicato, and Kim [14] have been investigating different forms of electron correlation in a multiconfiguration Dirac-Fock model, and their preliminary lifetime calculations are indeed strongly dependent on the configurations used. However, it is promising that some of their configurations yield lifetimes exceeding the experimental values by only 20–30 %. More on this apparent progress will be forthcoming.

Perhaps more relevant to the present discussion is the Z dependence found by different investigators. Katori and Shimizu's trap-loss measurements suggest very little difference between the Ar and Kr lifetimes, and their results lie surprisingly close to the Xe value of Ref. [4]. However, when the Xe result and our present measurement are considered together, it appears that these two VUV-photon-counting measurements confirm the theoretical expectation of longer lifetimes for higher Z. In fact, we find a Kr:Xe lifetime ratio that is quite close to what is predicted by the calculations of Desclaux, Indelicato, and Kim. This outcome tends to foster confidence in the general trend predicted by theory.

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