Lifetime measurements using the cascade-photon-coincidence technique with a sputtered-atom source

S. D. Rosner,* R. A. Holt, and T. J. Scholl
Physics Department, University of Western Ontario, London, Ontario, Canada N6A 3K7
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We have developed an interesting and widely applicable technique for measuring lifetimes of low-lying levels of neutral atoms and their first few ionization states. The cascade-photon-coincidence method has been extended by using a light source consisting of excited atoms sputtered from a solid by 10-keV argon ions. We have measured the lifetimes of the $4p\,^2P^o_{3/2}$ state of Ca II, the $6p\,^2P^o_{3/2}$ state of Ba II, and the $4p\,^2P^o_{3/2}$ state of Al III. These measurements are in good agreement with earlier work. [S1050-2947(97)04805-1]

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I. INTRODUCTION

The measurement of radiative lifetimes and oscillator strengths is important in several fields of physics [1–7]. In astrophysics in particular, the extraction of interstellar column densities from experimental absorption line profiles requires accurate resonance oscillator strengths. Until fairly recently, ground-based optical-absorption studies of the interstellar medium were limited to a small number of elemental ions of low cosmic abundance due to atmospheric absorption. The Goddard High Resolution Spectrograph aboard the Hubble Space Telescope has made possible the detection of many abundant ions through observation of ultraviolet absorptions of their resonance lines [8]. These spectra are of such excellent quality that the derived abundances are now in many cases limited by the knowledge of the oscillator strengths.

The cascade-photon-coincidence (CPC) technique, used extensively in nuclear physics, was originally applied to atomic lifetime measurement by Brannen *et al.* [9]. This precision technique involves the direct measurement of the time interval between the spontaneous emission of two sequential photons in an atomic cascade. The emission of the first (''start'') photon signals an atomic transition to a state whose lifetime is to be measured; a second (''stop'') photon signals the decay of that state. The histogram of these time intervals is a convolution of the instrumental time response of the apparatus with an exponentially decaying curve whose decay constant is the desired lifetime.

In this paper, we will show that the combination of the CPC method with a sputtered-ion light source provides an excellent means of measuring atomic lifetimes of low-lying states of astrophysically important ions. Virtually any element that can be incorporated into a solid material can be sputtered, producing abundant yields of ions and neutrals that are non-selectively excited to low-lying states. This is a significant advantage over beam-laser methods, where it is often quite difficult to form a beam of ions or fast neutrals from refractory materials using a readily available ion source. In the case of refractory metals, the useful operating time of such a high-temperature ion source is seriously limited by the coating of insulators inside the source.

Nonselective excitation is a serious problem for beam-foil experiments, causing systematic errors in lifetime measurements due to radiative cascading from higher levels. However, by its very design, the CPC technique is free from systematic errors caused by cascades from higher states and thus does not require selective excitation to the state of interest: an extremely high degree of selectivity is instead provided by the start and stop photon detection. Furthermore, the sputtering light source is extremely bright and can be made quite small, which is crucial for light collection efficiency in a coincidence experiment. The cascades that can be investigated with this technique are limited in wavelength range only by transmission through the optical system and the spectral response of the detectors. The prototypical experiments described below have shown that $\sim 1\%$ accuracy can be obtained for lifetimes under ~ 20 ns, given a correctly designed optical collection system. The present work also represents the first CPC experiment employing two monochromators for wavelength discrimination instead of interference filters [10].

II. EXPERIMENT

The excited neutral atoms and ions were produced by sputtering material from a solid target with a 10-keV argon-ion beam. The beam of Ar^+ ions was produced in a Colutron ion source in a conventional accelerator [11]. The mass-filtered beam was electrostatically deflected and focused through two circular apertures to be normally incident on the sputtering target. The diameter of the ion beam on the target was determined by the final 0.5-mm-diameter aperture located approximately 3 cm upstream from the target. The target and both beam-defining apertures were fixed to a translation stage which permits movement along the ion beam axis. The precise location of the target relative to the primary focus of the light-collection system was determined by optimizing the product of the start and stop counting rates. Typical argon-ion sputtering currents were ~ 100 nA. The vacuum in the sputtering chamber was 5×10^{-7} Torr.

The sputtering targets of calcium and barium were machined from metal pieces that were $>99\%$ pure, while the aluminum target was made from ordinary machine-grade

*Electronic address: rosner@uwo.ca

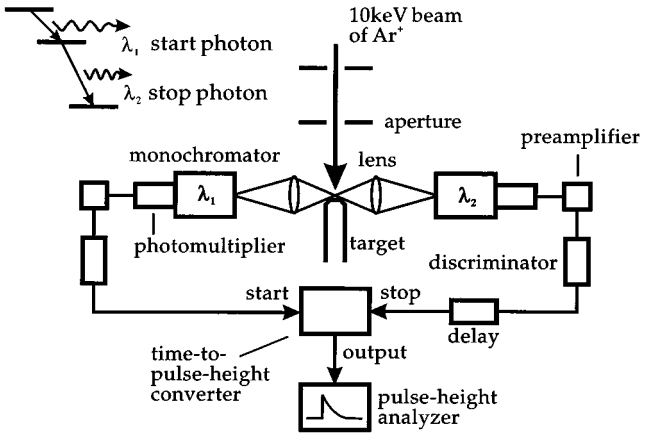

FIG. 1. Schematic diagram of the experimental apparatus including the timing electronics. The source of excited atoms is formed by sputtering neutrals and ions from a solid target with a fast beam of argon ions.

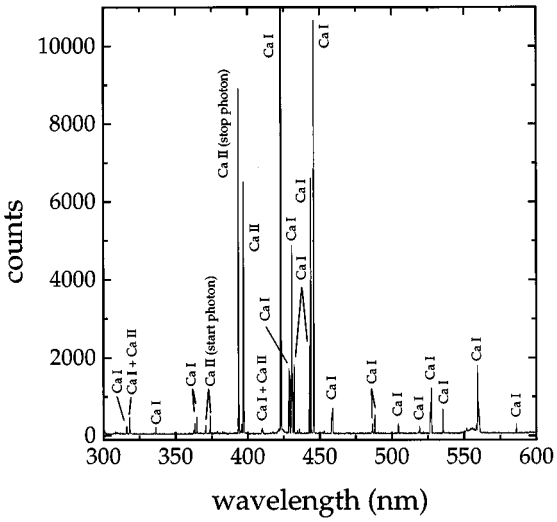

FIG. 2. Optical spectrum of spontaneous emission from excited calcium neutrals and ions sputtered from a solid target by 10-keV argon ions. The Ca I emission at 422.7 nm is approximately four times more intense than the second most intense line in this spectrum whose wavelength is ~445.5 nm.

aluminum. To reduce vignetting of the spontaneous emission by the target itself, the beam-facing side of a sputtering target was machined to a ~50° full-angle cone which was subsequently truncated to produce a 2-mm-diameter surface perpendicular to the ion beam. For aluminum, this surface was further enhanced by ruling parallel corrugations 0.1 mm deep every 0.1 mm to increase the yield of sputtered ions by using a 45° angle of incidence. The targets were grounded through an electrometer to permit monitoring of the incident argon ion current.

The sputtering process produces several sputtered particles for each incident ion. Typically about 10% of the sputtered particles are ionized and some fraction of those are in excited states, depending upon the angle of incidence, energy, and mass of the incident ion and the oxidation of the target surface [12]. We observe spontaneous emission from excited sputtered particles at distances up to ~1 mm from the target surface. Neglecting repopulation of the intermediate state by cascades from long-lived higher states, and assuming that the light curve is emitted over three lifetimes, one calculates an upper limit for the ion energy of the $4p\,^2P^o_{3/2}$ state of Ca II to be ~500 eV. To avoid systematic errors from atoms escaping the viewing region before decaying, the light collection system must be capable of collecting light from the entire spatial distribution of light-emitting atoms.

The spontaneous emission was gathered by two $f/1.4$ lens systems facing each other and viewing at right angles to the ion beam. The lenses imaged the source onto the entrance slits of separate monochromators (see Fig. 1). The viewing regions of both optical systems were carefully aligned to within 0.1 mm by substituting a small light bulb for the sputtering target. The theoretical lateral magnification for the lens system in the paraxial approximation was 2.3. The two $f/3.8$ monochromators each had three selectable gratings and adjustable slits. For spectral surveys (see Fig. 2), the monochromator slits were narrowed to provide 0.375-nm resolution. Further higher-resolution spectra were acquired in the vicinity of the start and stop photon wavelengths to check for interfering cascades (undesired cascades whose start and stop wavelengths were both within the passbands of the corresponding monochromator). Typically, coincidence data were gathered with the slit widths set to the maximum of 3 mm, which corresponded to 3-nm resolution.

It is interesting to note that little or no light is observed from the incident argon ions. Since they have a range of approximately 100 nm in these materials, it is likely that ions which may be excited as they encounter the surface are quenched before they can radiate. The spectrum (see Fig. 2) is dominated by narrow line emissions from target atoms, indicating that very little light comes from excited bulk material. The spectra also show that, for these target materials and experimental parameters, only the first few levels above the ground state for each charge state were populated by the sputtering process. Although this precludes the measurement of lifetimes of higher atomic states, these states are not typically populated in the interstellar medium. In fact, most interstellar atoms and ions are observed by absorptions from the ground state to their first excited state. The modest level of excitation also means that the spectra are relatively sparse, and a chosen radiative cascade can be easily isolated with monochromator slits fully opened to maximize optical efficiency over the entire source volume.

The photons transmitted by the monochromators were detected by Philips XP2020Q photomultiplier tubes. Their output pulses were preamplified (Ortec model 9301), discriminated in constant fraction mode (Ortec model 473A), and then used to start and stop the time-to-pulse-height converter (TPHC) (Ortec model 467). To ensure that the TPHC was operated in its most linear region and to provide a region of random background for curve fitting, ''time zero''was moved to the center of the TPHC range by inserting a fixed time delay (Ortec 425A) in the stop channel. Finally, the TPHC pulses were digitized and stored by a pulse-height analyzer (PHA)(Tracor-Northern model 1750) producing a histogram of time delays. The temporal response of the detectors and timing electronics was measured by placing a Čerenkov light source, consisting of 100 μCi of ^{90}Sr/^{90}Y in

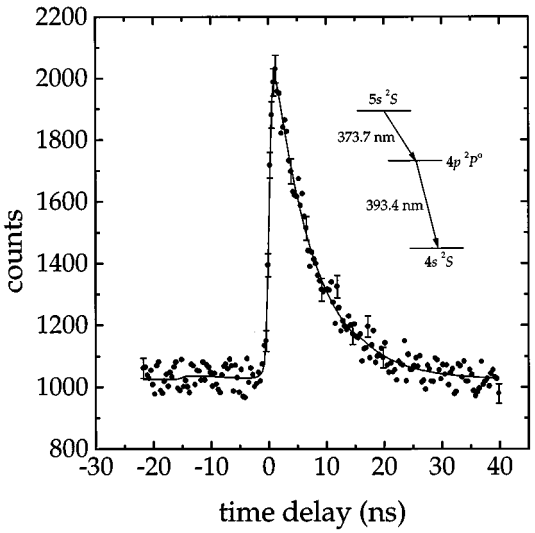

FIG. 3. The time-delay spectrum for the measurement of the lifetime of the $4p\,^2P^o_{3/2}$ state of Ca II. The least-squares fit of the theoretical decay curve convolved with the instrumental response of the timing apparatus is shown as the solid line. Representative error bars are shown only at each end of the histogram and scattered over the decay curve for clarity. The lifetime was determined to be 6.94(18) ns and the χ^2 per degree of freedom for the fit was 1.09.

a Lucite block, at the focus of the optical system. The ~1-ns full width at half maximum timing resolution was due mainly to the photomultiplier response function. The unusual-looking bumps in the least-squares fit to the time-delay histograms (see Figs. 3, 4, and 5) are due to the shape of this response curve and are a result of elastic scattering of the photoelectrons at the first dynode of the photomultipliers [13]. Calibration of the TPHC-PHA timing system has been

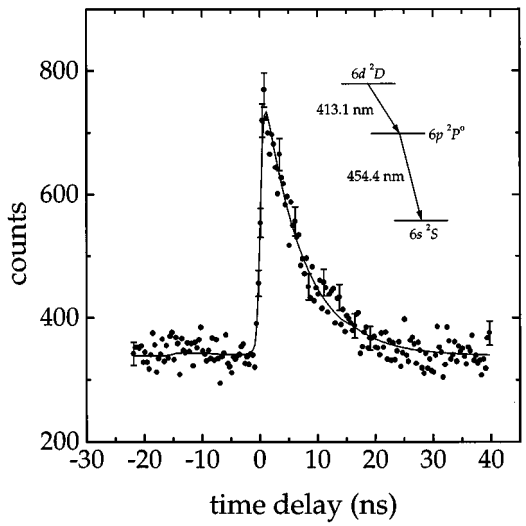

FIG. 4. The time-delay spectrum for the measurement of the lifetime of the $6p\,^2P^o_{3/2}$ state of Ba II. The least-squares fit of the theoretical decay curve convolved with the instrumental response of the timing apparatus is shown as the solid line. Representative error bars are shown only at each end of the histogram and scattered over the decay curve for clarity. The lifetime was determined to be 6.57(24) ns and the χ^2 per degree of freedom for the fit was 1.09.

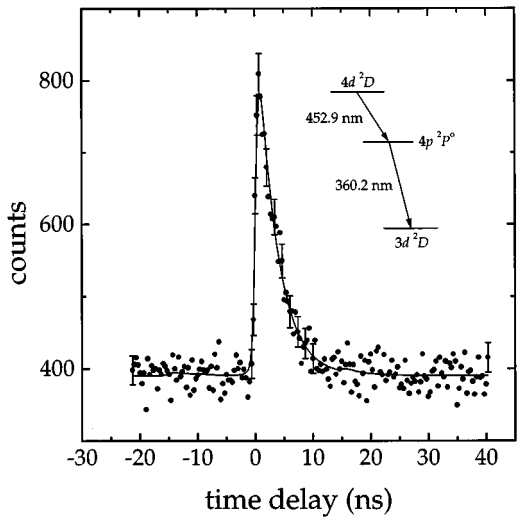

FIG. 5. The time-delay spectrum for the measurement of the lifetime of the $4p\,^2P^o_{3/2}$ state of Al III. The least-squares fit of the theoretical decay curve convolved with the instrumental response of the timing apparatus is shown as the solid line. Representative error bars are shown only at each end of the histogram and scattered over the decay curve for clarity. The lifetime was determined to be 3.24(16) ns and the χ^2 per degree of freedom for the fit was 0.83.

described in detail previously [14].

In addition to the exponential decay curve of true coincidences, there is a background of random coincidences which occurs when the start and stop pulses arise from either different atoms or from dark counts in the photomultipliers and hence are temporally uncorrelated. The ratio of true coincidences to random coincidences can be manipulated by varying the intensity of the source of excited atoms. Since the random coincidence rate varies quadradically with the source intensity, while the true coincidence rate varies linearly, decreasing the source intensity increases the signal-to-background (S/B) ratio at the expense of longer data collection times. A reasonable compromise in source intensity was chosen to provide a S/B ratio of ~1.

III. RESULTS AND DISCUSSION

Experimental parameters for the lifetime measurements of the $4p\,^2P^o_{3/2}$ state of Ca II, the $6p\,^2P^o_{3/2}$ state of Ba II, and the $4p\,^2P^o_{3/2}$ state of Al III are summarized in Table I. The corresponding time-delay coincidence histograms and the least-squares fit to them are shown in Figs. 3, 4, and 5 respectively. The time-delay histograms were analyzed using a convolution of the measured instrumental response function with the expected time-delay spectrum. Since only the first stop signal following any start signal is recorded, there is a bias against detecting long-time delays when the stop rate is high enough such that the probability of more than one stop event over the time span of the histogram is non-negligible. To correct for this systematic effect known as ''droop,'' the experimental decay curves were first divided by $\exp(-R_{\rm stop}t)$, where $R_{\rm stop}$ is the observed collection rate of stop photons. This correction factor represents the Poisson-distribution probability that no stop event will occur between times 0 and t. It had the largest effect for the Ca measure-

TABLE I. Summary of experimental parameters for measurements of lifetimes in Ca II, Ba II, and Al III using the cascade-photon-coincidence technique.

State	Atom	Start λ (nm)	Stop λ (nm)	Start rate (kHz)	Stop rate (kHz)	Acquisition time (h)	Coincidence rate (Hz)
$4p\,^2P^o_{3/2}$	Ca II	373.7	393.4	1.7	21.0	22.6	0.31
$6p\,^2P^o_{3/2}$	Ba II	413.1	454.4	1.8	7.5	22.1	0.12
$4p\,^2P^o_{3/2}$	Al III	452.9	360.2	2.6	4.5	29.4	0.053

ment; even there it led to a negligible shift of the lifetime by $+0.006$ ns.

The observed time-delay histograms for a coincidence experiment can be distorted in different ways by an improperly designed optical collection system. This is particularly true when the atoms being measured have an appreciable velocity, creating a more extended light source. If the optical systems monitoring start and stop photons are not well aligned, it is possible that the apparatus will preferentially detect stop photons for either short- or long-time delays. Even if the alignment is perfect, another possibility is that the ion velocity is so large that many ions escape the viewing region before emitting a stop photon, leading to an observed lifetime which is systematically too short. The first potential systematic error was prevented by careful alignment of the two optical systems using a point light source as described above. The spatial distribution of spontaneous emission at the stop wavelength was recorded by translating the sputtering target along the ion beam direction in front of the light collection systems. In calcium, for example, the emission rapidly decreased to a negligible amount over a 1-mm extent. The 3-mm-wide monochromator slits viewed a 1.30-mm region, taking into account the magnification of the optical system. Thus the monochromators were able to collect the spontaneous emission from the entire source volume.

To explore any other systematic effect which could arise from the size, shape, and positioning of the sputtering target we modeled the experiment using a highly realistic Monte Carlo simulation. This simulation traced rays for the spontaneous emission at both the start and stop wavelengths from moving sputtered ions to the entrance slits of the monochromators. We assumed that the sputtered ions had velocities given by a Thompson energy distribution [15], with a surface binding energy of 12 eV and a maximum ion energy of 3 keV. This last parameter is obviously an overestimate in light of the previous discussion. The sputtered ions were given a $\cos\theta$ distribution cylindrically symmetric with respect to the surface normal. Least-square fits to the synthetic time-delay histograms showed that any effect of the ion velocity on the observed lifetime is below the 0.5% level.

There are a number of collisional effects which could in principle create systematic errors in the lifetime. Collisions between sputtered atoms and the incident fast Ar^+ beam are highly improbable over the time intervals of a few lifetimes. Typical Ar^+ beam parameters and a cross section of 10^{-15} cm^2 yield a collision probability of $\sim 10^{-7}$ in the time of transit over the viewing region. We also examined the effect of simultaneous production of more than one excited sputtered ion by an incident fast argon ion. Since the sputtering event lasts only ~ 100 fs, photons from different ions would be correlated in time when viewed on a nanosecond time scale. If this process were present, we would expect to see a coincidence peak in the time-delay spectrum when both channels were tuned to the same photon wavelength, but none was seen in a separate experiment undertaken to study this possibility.

During the course of these measurements we observed that the apparent measured lifetime increased noticeably at extremely high incident ion currents; an example is shown in Fig. 6. One of the mechanisms considered as an explanation was radiation trapping, in which the stop photon is subsequently reabsorbed by another atom, which then emits another photon at a later time. If the probability for such reabsorption is p, then the apparent lifetime τ_a is given by $\tau_a = \tau_{\text{true}}/(1-p)$ [16]. Since p depends on the density of atoms in the final state of the cascade, radiation trapping will be significant typically when the final state is a ground or metastable state, as is true for the Ca II and Ba II cascades studied here. A calculation of p based on the resonance-line oscillator strength and an estimated ground-state density of sputtered ions shows $p < 1\times 10^{-5}$. Also, the expected linearity of τ_a^{-1} with p was not observed in measurements on Ca II. Our experience to date suggests that reducing the sputtering current produces a rapid convergence to the true lifetime. Since the source is extremely bright, the resulting coincidence rate is still sufficient to permit data collection over a reasonable time period. Furthermore, the reduced start and stop rates increase the observed S/B ratio and virtually eliminate the droop correction. Nevertheless, further study of the

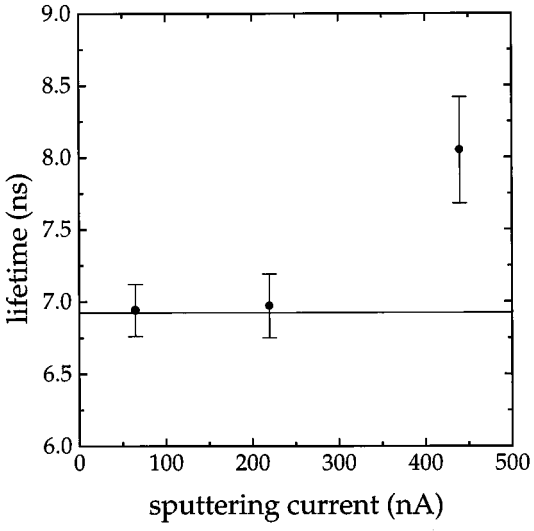

FIG. 6. Dependence of measured lifetime of the $4p\,^2P^o_{3/2}$ state of Ca II on the incident Ar^+-ion beam sputtering current. The horizontal line at 6.924 ns indicates the value obtained in Ref. [17].

TABLE II. Comparison of experimental measurements of lifetimes in Ca II, Ba II, and Al III. BL, beam-laser method; BF, beam-foil method; CPC, cascade-photon-coincidence method.

Author(s)	Reference	Method	Lifetime (ns)
	$4p\,^2P^o_{3/2}$ state of Ca II		
Gosselin, Pinnington, and Ansbacher	[18]	BL	6.87(6)
Jin and Church	[17]	BL	6.924(19)
This work		CPC	6.94(18)
	$6p\,^2P^o_{3/2}$ state of Ba II		
Andrä	[19]	BL	6.312(16)
Winter and Gaillard	[20]	BL	6.31(5)
This work		CPC	6.57(24)
	$4p\,^2P^o_{3/2}$ state of Al III		
Anderson, Jessen, and Sørenson	[21]	BF	3.4(3)
Coetzer, Mostert, and Van der Westhuizen	[22]	BF	3.0
This work		CPC	3.24(16)

sputtering-current effect is clearly warranted, and will be undertaken with an apparatus whose data-collection rate will be substantially improved.

The results of least-squares fitting to the decay histograms are compared to the most precise previous measurements in Table II. Measurements using the CPC technique are in good agreement with previous high precision work using the beam-laser method for both the $4p\,^2P^o_{3/2}$ state of Ca II and the $6p\,^2P^o_{3/2}$ state of Ba II, but are not nearly as precise. Previous to this work, the lifetime of the $4p\,^2P^o_{3/2}$ state of Al III had been measured only by beam-foil methods that had been corrected for radiative cascading. The present measurement is more precise than the beam-foil measurements and is in good agreement.

This experiment is a demonstration that the cascade-photon-coincidence technique works well with a sputtered source of excited atoms. It also demonstrates the use of monochromators for wavelength discrimination, which eliminates the need for custom interference filters for each cascade and extends the technique into the UV region where narrow-band interference filters are more difficult to fabricate. We have already considered experimental improvements to increase the optical efficiency of the apparatus greatly, and are planning construction of a dedicated apparatus for lifetime measurement with this technique. The first important change will be to build a catoptric light collection system consisting of two opposed coaxial ellipsoidal reflectors with common first foci. The sputtering target will be introduced through a hole in the ellipsoid pair at right angles to the optical axis. Spontaneous emission from excited sputtered atoms will then be collected by the ellipsoids, and reflected directly into the entrance slit of either the start or stop monochromator. This should extend the wavelength capability of this technique down to the vacuum cutoff. A low f number monochromator for the stop channel will be used to accept the larger solid angle of the light collected by the ellipsoid. Monte Carlo simulations of this achromatic optical system and faster monochromator confirm that accurate lifetime measurements will be possible at the $\sim 1\%$ level with an increase in the coincidence rate of over two orders of magnitude without the use of interference filters.

IV. CONCLUSIONS

We have measured the lifetimes of the $4p\,^2P^o_{3/2}$ state of Ca II, the $6p\,^2P^o_{3/2}$ state of Ba II, and the $4p\,^2P^o_{3/2}$ state of Al III using the CPC technique with a source of excited atoms sputtered from a solid. The results are in good agreement with previous higher precision work using the beam-laser method for both Ca II and Ba II. The lifetime measurement for Al III is also in agreement with earlier work, and is the most precise measurement of the lifetime of that state. A second-generation apparatus with better optical efficiency based on this work has been described that will be used for lifetime measurement of low-lying states of astrophysically interesting atoms and ions.

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