Lifetime Measurements on Long-Lived Levels of Confined Highly Charged Ions

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A technique based on the injection of highly charged ions from new low-energy ion sources into a Kingdon ion trap has been used to measure millisecond or longer lifetimes τ of metastable levels in ground terms of the ions. The technique has been initially applied to ions of argon, with the results $\tau(Ar^{10+},2p^4\,{}^3P_1)=14.8(1.1)$ ms, $\tau(Ar^{9+},2p^5\,{}^2P_{1/2})=8.53(0.24)$ ms, and $\tau(Ar^{2+},3p^4\,{}^1S_0)=159.7(9.7)$ ms.

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For ions in a variety of charge states, lifetimes of both allowed and "forbidden" transitions are used to compute electron densities and temperatures in astrophysical and laboratory plasmas [1]. Abundances of elements in the solar corona are determined from observed magnetic dipole transitions in gound configurations [2]. Prominent lines of high charge states of, e.g., O, Si, Fe, Ni, and Ar provide useful diagnostics in the solar atmosphere [1–3]. Lifetimes of metastable levels of highly charged ions have been extensively calculated [4–6], since this information has not been available experimentally. The accuracies of most such calculations are difficult to estimate, and recourse to experiment is useful to evaluate the quality of the theory. Relativistic many-body perturbation theory (MBPT) calculations permit estimates of accuracy based on the order of perturbation [7], but recent MBPT lifetime calculations have differed significantly in some cases from measurement results on singly charged ions [8]. Experimental data on ions in which electron correlation and relativistic effects are important are desired to enable further evaluation of these and other calculational techniques. The first experimental results on levels of highly charged ions with millisecond or longer lifetimes are presented here.

Many ions in high charge states have metastable levels with lifetimes exceeding a millisecond, and decays with wavelengths lying in the ultraviolet to near infrared range [9]. The ion containment technique is ideal for measurement of the lifetimes of such long-lived states [10], but no measurements on charge states higher than 3+ had been completed prior to this work, due to difficulties in producing sufficient ions with higher charge inside the trap. We report the successful injection and capture of highly charged low-energy ions in metastable levels from an external ion source into a Kingdon ion trap, followed by metastable level lifetime measurements. Results for Ar^{10+}, Ar^{9+}, and Ar^{2+} are discussed, although these methods are applicable to metallic as well as gaseous ions, in any charge state that can be produced in continuous or pulsed beams from low-energy (kV) ion sources. Such sources include the Electron Cyclotron Resonance Ion Source (ECRIS) [11] used in these measurements, as well as the Electron Beam Ion Source (EBIS) [12] and the Electron Beam Ion Trap (EBIT) [13], when used as a source [14]. A complementary technique based on ions stored in EBIT useful for levels with microsecond lifetimes has been developed subsequent to our work [15].

Figure 1 diagrams our basic apparatus configuration. A 15 cm long Kingdon ion trap consisting of a 10 cm diam aluminum cylinder with separate plane end electrodes and a 0.125 mm diam coaxial tungsten wire confined the ions. The midplane of the cylinder was perforated by four 1.9 cm diam apertures. Two in-line apertures passed an ion beam radially through the trap to a Faraday cup (FC), while the apertures at right angles opened respectively onto a microchannel plate (MCP) ion detector and the quartz optics of a photon-collection system. Initially the trap cylinder and central wire were held at a potential close to the extraction voltage of the ion source, typically set to 3.5 kV for reasons of trap voltage stability. Multicharged ions produced in the ECRIS discharge, magnetically separated into a beam with a single charge state, were electrostatically focused through a grounded cylinder, which acted as a lens, into the trap. The consequent rapid deceleration of the ions increased their density and led to defocusing consistent with phase space conservation. When the trap central wire was rapidly (≈ 100 ns) pulsed to 0 V, those ions within the trap having sufficient angular momentum relative to the central wire were confined while the wire potential remained low. Potentials on the end caps exceeding the cylinder voltage held the ions near the trap midplane. Long-lived

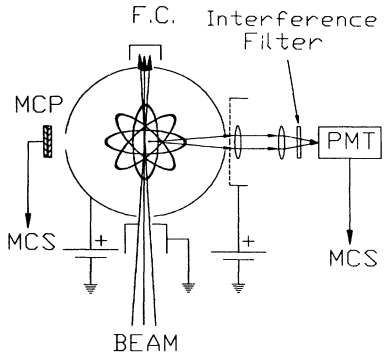

FIG. 1. Apparatus configuration, with the Kingdon trap shown in cross section at the midplane.

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metastable states produced in the ion source emitted photons during the ion storage interval, which were collected by the optical system, selected in wavelength by an interference filter, and counted versus storage time using a photomultiplier tube (PMT) and multichannel scalar (MCS). After predetermined ion storage times, the ions were dumped by allowing the central wire potential to rise with a time constant of milliseconds to the cylinder potential. A fraction of the ejected ions was counted versus storage time using the MCP. The ion beam was deflected using up-beam plates during the ion confinement interval.

The ground terms of three argon ion charge states are diagramed in Fig. 2. The Ar^{9+}, $2s^2 2p^5\ ^2P_{1/2}$ level radiates a magnetic dipole ($M1$) or with lower probability an electric quadrupole ($E2$) photon with wavelength $\lambda = 553$ nm to reach the $^2P_{3/2}$ ground state. Using the multiconfiguration Dirac-Fock (MCDF) theory [4], the lifetime of the $^2P_{1/2}$ level has been calculated to be 9.43 ms. Accuracy is unlikely to exceed 10%, since only $n=2$ configurations were included. Experimentally an optical

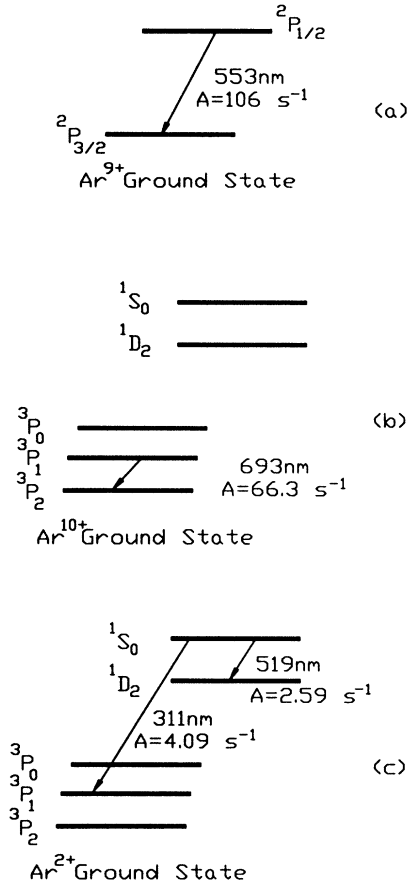

FIG. 2. Ground term level diagrams with wavelengths of measurement transitions, for (a) Ar^{9+}, (b) Ar^{10+}, and (c) Ar^{2+}.

decay was observed when Ar^{9+} was stored, which was fitted well by two exponentials. A rapid initial decrease in intensity was also observed in separate measurements of transitions of other charge states, and is attributed to transient effects associated with ion capture into the trap. The time constant of the longer-lived decay component decreased when the residual pressure in the vacuum system was increased from its base near 3×10^{-9} Torr, as expected for collisional quenching. This quenching is interpreted as any loss mechanism for the metastable ions due to collisions. Stern-Volmer plots of the reciprocal of the fitted time constant of this component, as a function of the pressure of either Ar or N_2 gas admitted to the target chamber in the 10^{-7} to 10^{-8} Torr range, have slopes which give the quenching rate coefficient due to that gas, and a zero pressure intercept (including residual gas effects) which can be interpreted as the reciprocal of the lifetime of the metastable state. The quenching rate coefficients were $k(Ar^{9+},Ar) = 8.5(0.5)\times10^{-8}$ cm^{3}s^{-1} and $k(Ar^{9+},N_2) = 7.3(0.4)\times10^{-8}$ cm^{3}s^{-1}, while the common zero-pressure intercept yielded a time constant of 8.36(0.12) ms, slightly lower than but in adequate agreement with the MCDF calculation.

The coupling of two holes in the L shell produces the ground term of Ar^{10+}, diagramed in Fig. 2(b). Only the $^3P_2$-$^3P_1$ $M1$ transition near 693 nm had a convenient wavelength for measurement. The deep uv cascade decays, $^3P_1$-$^1S_0$ and $^3P_1$-$^1D_2$, were calculated to occur within a few milliseconds [9], contributing solely to the initial transient observed in the $^3P_2$-$^3P_1$ decay. On the other hand, the solution of the rate equations, assuming an initial statistical population of the ground term levels, indicated that the long-lived $^3P_1$-$^3P_0$ decay should have an amplitude comparable to the statistical fluctuations of the signal. Experimentally, the data were again well fitted by two exponentials plus a constant background. A Stern-Volmer plot for the longer-lived component of the optical decay (see Fig. 3) yielded a zero-pressure inter-

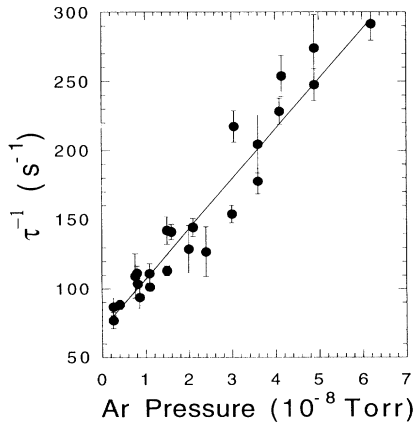

FIG. 3. Plot of reciprocals of measured lifetimes τ_m^{-1} vs pressure for the Ar^{10+} $^3P_2$-$^3P_1$ transition near 693 nm.

cept corresponding to a lifetime of 14.3 ms, close to the MCDF calculation $\tau(Ar^{10+},3p^4\,{}^3P_1)=15.08$ ms. The quenching rate coefficient $k(Ar^{10+},Ar)=10.1(0.6)\times10^{-8}\,cm^3s^{-1}$.

To further test the accuracy and precision of the technique, measurements on the $Ar^{2+},3p^4\,{}^1S_0$ decays [see Fig. 2(c)] were carried out. Two previous measurements on this low charge state had been completed, also in Kingdon traps, using electron impact ionization to produce the metastable ions. In each case, the stronger $^3P_1$-$^1S_0$ $M1$ branch near 311 nm was studied. The resulting decay time constants were $\tau(Ar^{2+},3p^4\,{}^1S_0)=109(32)$ ms [16] and $\tau(Ar^{2+},3p^4\,{}^1S_0)=133(24)$ ms [17]. Our measurements at the 311 nm wavelength agreed with these and with our more extensive measurements on the $^1D_2$-$^1S_0$ $E2$ decay near 519 nm, resulting in a decay time constant of 121.3(3.4) ms, in excellent agreement with earlier results, but about 25% below the best theoretical calculations [5,6]. The quenching rate coefficients in Ar and N_2 gas, $k(Ar^{2+},Ar)=4.2(1.1)\times10^{-9}$ cm^3s^{-1} and $k(Ar^{2+},\ N_2)=4.5(0.9)\times10^{-9}$ cm^3s^{-1}, were similar, and well below the rates for the higher charge states, as anticipated.

The storage time constant for ions in the trap was also obtained by plotting the time dependence of the MCP signal produced by ions ejected from the trap at the end of the storage time. This signal was observed to decrease exponentially with time for both Ar^{2+} and Ar^{9+} ions at constant pressure, with a 4.9 times more rapid rate of ion loss for the higher charge state. The scaling is approximately proportional to the charge, as expected for these low-energy collisions. The storage time for each charge state decreased as the pressure in the target chamber was increased, as expected for ion storage limited by ion-neutral charge-exchange collisions. Extrapolation to zero pressure in each case resulted in a finite ion storage time constant, 505(39) ms for Ar^{2+} and 427(62) ms for Ar^{9+}, which did not depend significantly on ion charge state. Measurements with different numbers of stored ions, to test for possible effects of ion-ion collisions, and with different trap operating voltages, to test for well-depth effects, did not produce any changes of significance. Consequently, the source of this apparent pressure-independent limitation on ion storage time has not been ascertained, although it may be related to the diameter of the central wire of the trap.

However, a finite ion storage time will affect the value of the lifetime of the metastable level obtained from a Stern-Volmer plot, since $\tau^{-1}=\tau_m^{-1}-\tau_{is}^{-1}$, where τ_m is the measured time constant obtained from the zero-pressure intercept, τ_{is} is the mean finite ion storage time constant, and τ is the natural lifetime of the level. These corrections to the data were carried out by direct subtraction, but since the source of a pressure-independent value of τ_{is} has not been definitely identified, these corrections are treated as systematic errors, with the random errors of the zero-pressure intercepts of the optical decay measurements and the finite storage time measurements added in quadrature to obtain the final random error estimates. Our corrected results for the mean lifetime measurements are summarized in Table I.

TABLE I. Lifetime measurement results with random and systematic error estimates. The systematic shift is due to pressure independent ion storage limitations.

Level	Lifetime (ms) Expt.	Theory
Ar^{10+}, $2p^4\,{}^3P_1$	$14.8(\pm1.1,-0.5)$	15.08[a]
Ar^{9+}, $2p^5\,{}^2P_{1/2}$	$8.53(\pm0.24,-0.17)$	9.43[a]
Ar^{2+}, $3p^4\,{}^1S_0$	$159.7(\pm9.7,-38.4)$	152.8[b]
		149.4[c]

[a]Reference [1].
[b]Reference [2].
[c]Reference [3].

Inclusion of the systematic error due to finite storage time had by far the largest effect (25%) on the Ar^{2+} optical decay, which had the longest mean lifetime. Experimental uncertainty is also dominated by the systematic error. The results of the measurements are in all cases in good to excellent agreement with available theory, assuming $\approx 10\%$ theoretical uncertainties.

In conclusion, we have presented the initial data obtained by a technique which is applicable to any ion charge state available with sufficient particle current ($\lesssim 1$ μA equivalent) from a low-energy ion source, and to metastable lifetimes longer than a few milliseconds. An upper limit on measurable time constants may be set if the limited zero-pressure ion storage time proves intractable. The technique is expected to permit lifetime measurements of isoelectronic states as a function of nuclear charge, and of all transitions between ground levels of a given term, by using different charge states for particular transitions. Determination of the effects of hyperfine interaction on certain metastable level lifetimes is also feasible.

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