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Lifetimes of the 7p levels in Hg II

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Measurements of the lifetimes for the $5d^{10}7p\ ^2P_{1/2}$ and $^2P_{3/2}$ levels in Hg II have been made using beam–foil excitation. The lifetimes were extracted both by exponential curve fitting of the individual decay curves and by joint analysis of the cascade-correlated decay curves using the ANDC method. The results are $\tau(J=\frac{1}{2}) = 14.5 \pm 1.0$ ns and $\tau(J=\frac{3}{2}) = 1.2 \pm 0.2$ ns. Theoretical computations are presented indicating that this large J dependence results from the combined influence of a near-lying cancellation node and CI-induced decay to the $5d^9 6s^2$ levels.

The lifetimes of the $5d^{10}7p\ ^2p_{1/2}$ and $^2P_{3/2}$ levels in Hg^+ are worthy of experimental study for a number of reasons. Their intrashell transitions to $5d^{10}7s\ ^2S_{1/2}$ give rise to the well-known He–Hg laser lines at 7944 and 6149 Å (see refs. [1,2], and references therein), and a knowledge of their transition probabilities and lifetimes would be useful in the study of this and other light sources that employ the mercury ion spectrum. Their resonance transitions to the ground state $5d^{10}6s\ ^2S_{1/2}$ at 893 and 923 Å are the second members of the principal Rydberg series in this ion, and one of the first members ($6s\ ^2S_{1/2}$–$6p\ ^2P_{1/2}$ at 1942 Å) has been observed in the spectra of chemically peculiar stars obtained with the Hubble space telescope [3]. The 923 Å line, being long of the Lyman limit, provides an absorption feature for use in the study of astrophysical abundances of chemical elements. One of the decay channels for $7p\ ^2P_{3/2}$ is to the $5d^9 6s^2\ ^2D_{5/2}$ level, the excitation energy and lifetime of which have been accurately measured [4] by Doppler-free two-photon absorption, and recommended as a possible optical frequency standard. Furthermore, the 7p levels possess a number of interesting subtleties that can limit the reliability of ab initio theoretical calculations, and make their accurate experimental determination especially important.

The first subtlety concerns the electric dipole transition integrals for the $5d^{10}6s\ ^2S_{1/2}$–$5d^{10}7p\ ^2P_{1/2,3/2}$ resonance transitions, which are very near to conditions of total cancellation [5] due to a cancellation node of the Cooper minimum type [6,7]. The conditions of cancellation can be characterized by the effective quantum numbers of the upper and lower levels. A near correspondence of the effective quantum numbers of one of the fine structure transitions to those at a cancellation node can cause J dependent anomalies, both in the lifetime ratio of the 7p levels and in the intensity ratio for the 6s–7p multiplet [5]. Where cancellation effects are large, the line strengths are very sensitive to small inaccuracies in the wave function and to the choice of gauge, making ab initio calculations very difficult. The second subtlety concerns configuration interaction (CI) with the $5d^9 6s6p\ ^2P$ levels, which lie very close to the $5d^{10}7p\ ^2P$ levels, and provide additional exit channels to the $5d^9 6s^2\ ^2D$ levels. Calculations described below indicate qualitatively that this interaction is strongly dependent upon J, affecting almost exclusively the $J=\frac{3}{2}$ level. However, these calculations re-

produce experimental energy levels poorly, and their quantitative reliability is questionable.

These J-selective cancellation and CI effects introduce complications into the measurement of both 7p lifetimes. The $^2P_{1/2}$ resonance decay channel is nearer the cancellation node and is also predicted to have a negligible CI decay channel, causing it to have an unusually long lifetime. The $^2P_{3/2}$ resonance channel is further from the cancellation node and is also predicted to have a significant CI decay channel, causing its lifetime to be considerably shortened.

In a pulsed-electron-beam delayed-coincidence measurement, Blagoev et al. [8] have shown that the 7p $^2P_{1/2}$ and $^2P_{3/2}$ levels do indeed exhibit substantially different decay curves. However, these measurements used an excitation pulse with an effective fall time of 2.3 ns, thus requiring that the first 8 ns of the decay curves be discarded, after which lifetime estimates were obtained by curve fitting to one and two exponential forms. In order to extract reliable lifetimes for these particular levels, the decay curves must be measured promptly after excitation, and there should be a detailed accounting for the effects of cascade repopulation.

We have performed a study of the lifetimes of the 7p levels in Hg II using the sharp cutoff provided by the sudden emergence from a foil by a fast ion beam, and have utilized the ANDC method [9] to jointly analyze the decay curves of the primary level and its significant cascades. It was necessary to use a relatively low energy beam in order to obtain a high charge state fraction of foil emergent singly charged ions. This was essential to obtain acceptable signal-to-noise ratios for radiative transitions from the less populous $n=7$ and 8 levels, and to avoid possible blending with the complicated and incompletely analyzed Hg IV and V spectra. Thus the measurement required great care, since the lifetime of one of the levels corresponds to a decay length that is of the same order of magnitude as the length of the beam that can be spatially resolved by the optical detection system, and the lifetime of the other level corresponds to a decay length over which the beam divergence could cause some fraction of the beam particles to escape from the viewing volume. In a lifetime study using beam–foil excitation the decay curve of the $^2P_{1/2}$ level will be affected by repopulation from both shorter- and longer-lived cascades, and by downstream escape of scattered ions from the viewing volume. In the case of the $^2P_{3/2}$ level, the exponential corresponding to its decay persists for only a short time after excitation.

Ions of $^{202}Hg^+$ were accelerated to an energy of 250 keV by the University of Toledo heavy ion accelerator and sent through a thin, self-supporting carbon foil (2.0–2.4 $\mu g/cm^2$). The radiation emitted by the foil-excited ions was dispersed with an Acton 1 m normal incidence vacuum monochromator and was detected with either a Channeltron ($\lambda=700$–1200 Å) or a solar-blind Hamamatsu detector ($\lambda=1200$–1900 Å). The entrance slit and the grating mask were configured so as to resolve a 1 mm segment of the beam. Decay curves were obtained by measuring the intensity of the spectral lines as a function of the distance along the beam from the foil. The foil was stepwise translated relative to the optical system by a precision drive. To compensate for variations in the beam intensity or possible changes of the foil properties during a measurement, decay curves were normalized to the intensity of undispersed light detected by a photomultiplier and fibre optic link that viewed the beam at a fixed distance downstream from the foil. The experiment was computer-controlled by an on-line data acquisition system [10]. Details of the accelerator and experimental setup are given in refs. [11,12].

The post foil velocity of the ions was determined by subtracting the energy loss in the foil from the terminal energy of the accelerator (calibrated to better than 1% by quantum beat measurements in neutral helium). The energy loss was specified using the computer code TRIM [13], which for 250 keV ions in carbon predicts electronic and nuclear stopping of 6.4 and 15.7 keV $cm^2/\mu g$. Because of its angular dependence the nuclear stopping was reduced to 30% of its total value, which is appropriate for ions emerging in the forward direction [14,15]. For 2.2 $\mu g/cm^2$ foils this energy loss is 23 ± 10 keV, leading to a post foil velocity of 0.47 ± 0.01 mm/ns.

At this velocity the 1 mm segment of beam viewed by the optical system corresponds to 2 ns, but the positional accuracy still permits the determination of lifetimes as short as 1 ns. (The ANDC method determines the primary lifetime from inherent differences in the exponential admixtures in the cascade and primary decay curves, and does not require

that the primary exponential term be resolved, or even appear in the measured decay curve.) The upper limit is set by the angular scattering of the beam particles by the foil, which can lead to the downstream escape of ions from the viewing volume. To investigate this effect, we performed calculations of the beam divergence using the tables of Sigmund and Winterbon [16], based on the theoretical formalism of Meyer [17]. These calculations indicated that 50% of the beam particles have a scattering angle of less than 6.5°. These calculations were incorporated into the uncertainty estimates. If the assertion by Andersen [18] that the profile of the light emitted downstream in narrower than the profile of scattered particles is valid here, then this calculation overestimates the loss.

Theoretical estimates of the transition probabilities were made using four methods. Semiempirial computations were made for transitions among the singly excited $5d^{10}nl$ levels for $n=6$–10 and $l=0$–2 using the semiempirical Coulomb approximation [19], providing estimates of the rates of both the decay and the cascade repopulation of the 7p levels. Ab initio calculations were also made using the Cowan suite [20] of multiconfiguration relativistic Hartree–Fock (MCRHF) programs, which provided estimates of the decay channels to $5d^96s^2$ that are opened by CI. In addition, ab initio calculations were made using the multiconfiguration Dirac–Fock package GRASP [21], using both the Coulomb and Babushkin gauges.

The results of these calculations for the decay of 7p are presented in table 1. Notice that these methods give quite different results for the 6s–7p transitions, which are heavily affected by cancellation. The Coulomb approximation and GRASP/Babushkin gauge calculations both correctly predict that the cancellation primarily affects the $J=\frac{1}{2}$–$\frac{1}{2}$ transition, although they differ quantitatively. The MCRHF and GRASP/Coulomb gauge calculations both incorrectly predict that the cancellation primarily affects the $J=\frac{1}{2}$–$\frac{3}{2}$ transition. Only the MCRHF code produced values for all decay branches. The Coulomb approximation code cannot include CI and could not be used to compute the branch to $5d^96s^2$, and difficulties in convergence were encountered with the GRASP code when the $5d^{10}6d$ configuration was included for a neutral or singly charged ion. Transition probabilities using these branches were summed in various ways to obtain lifetime estimates listed in table 1.

The decay curves were analyzed both by multiexponential fitting [22], and by the ANDC method [23]. The latter method determines the lifetime through a joint analysis that renormalizes the intensities of the primary and cascade decay curves to the slope of the primary decay curve in a manner that is governed by the population equation. Since the ANDC method internally determines their relative normalizations, the decay curves can be measured in any convenient branch. The ultraviolet branches 6p–*n*s and 6p–*n*d are expected (because of λ^3 factors) to have higher transition probabilities than the visible and infrared 7p–*n*s and 7p–*n*d branches that produce the direct cascades. All measurements were therefore carried out in the ultraviolet wavelength region.

To examine the relative intensities of the primary and cascade transitions, and possible blending with transitions from other charge states of Hg, survey spectra in the region $\lambda=700$–2500 Å were obtained at a point immediately downstream from the foil. All of the lines in this region that correspond to low-lying transitions in Hg II and to the strongest transitions in Hg III were identified. As has been observed in other studies of foil-excited low-energy heavy ions [12], lines corresponding to higher lying levels in these ions were weak or absent. The spectra showed that the Hg II 6s–7p resonance transitions (893 and 923 Å) had corresponding peak intensities that differed by nearly two orders of magnitude (in sharp contrast to the *LS* coupling value 2:1). In order to account for cascade repopulation of the 7p levels, searches were made for transitions from higher lying *n*s and *n*d levels. These revealed measurable intensities for the two branches of the 6p–8s decay (1430 and 1645 Å) and for the 6p–7d transitions (1354 and 1539 Å, the latter blended). There was no significant intensity observed in the regions of the 6p–9s and 6p–8d and higher lying transitions.

Figure 1 shows sample decay curves of the $5p^{10}7p$ ^2P_J levels, clearly indicating their strong dependence upon *J*. To compare with previous results, a preliminary data reduction was made using multiexponential curve fitting [22]. The parameters obtained from a two exponential fit to the $^2P_{1/2}$ decay curves agreed

Table 1
Theoretical calculations for the transition probabilities and lifetimes 7p in Hg^+, compared to measured lifetimes.

Transition	J–J'	λ (Å)	A (ns^{-1})	J–J'	λ (Å)	A (ns^{-1})
$6s\,^2S_J$–$7p\,^2P_{J'}$	$\frac{1}{2}$–$\frac{1}{2}$	923	0.4421 [a]	$\frac{1}{2}$–$\frac{3}{2}$	893	0.0146 [a]
			0.1965 [b]			0.0036 [b]
			0.0006 [c]			0.1076 [c]
			0.0419 [d]			0.2149 [d]
$7s\,^2S_J$–$7p\,^2P_{J'}$	$\frac{1}{2}$–$\frac{1}{2}$	7947	0.0422 [a]	$\frac{1}{2}$–$\frac{3}{2}$	6152	0.0315 [a]
			0.0365 [b]			0.0417 [b]
			0.0395 [c]			0.0453 [c]
			0.0482 [d]			0.0859 [d]
$6d\,^2D_J$–$7p\,^2P_{J'}$	$\frac{3}{2}$–$\frac{1}{2}$	30166	0.0027 [a]	$\frac{5}{2}$–$\frac{3}{2}$	15559	0.0021 [a]
			0.0013 [d]			0.0075 [d]
				$\frac{3}{2}$–$\frac{3}{2}$	14312	0.0003 [a]
						0.0010 [d]
$6s^2\,^2D_J$–$7p\,^2P_{J'}$	$\frac{3}{2}$–$\frac{1}{2}$	1732	0.0019 [a]	$\frac{5}{2}$–$\frac{3}{2}$	1308	0.2718 [a]
			0.0111 [b]			0.2829 [b]
			0.0037 [c]			0.3058 [c]
				$\frac{3}{2}$–$\frac{3}{2}$	1628	0.0002 [a]
						0.0027 [b]
						0.0013 [c]
calculated lifetime (ns)			2.05 [a]			3.12 [a]
			4.10 [b]			3.02 [b]
			22.86 [c]			2.17 [c]
			10.6 [d,e]			1.67 [d,e]
experimental lifetime (ns)			14.5 ± 1.0 [f]			1.2 ± 0.2 [f]
			18.8 ± 1.2 [g]			3.1 ± 0.2 [g]

[a] MCRHF [24]. [b] GRASP/Coulomb gauge [25]. [c] GRASP/Babushkin gauge [25]. [d] Coulomb approximation [23].
[e] Uses the average of a and c for the $5d^96s^2$ branches. [f] Beam–foil excitation, this work. [g] Pulsed electron beam excitation [8].

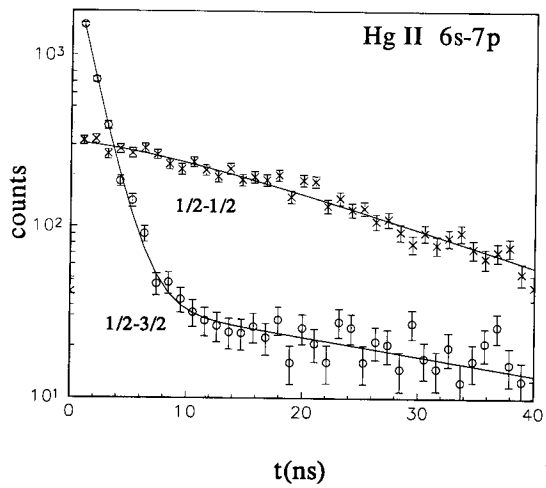

Fig. 1. Sample decay curves for the $7p\,^2P_{1/2}$ (×) and $7p\,^2P_{3/2}$ (○) levels. The data are shown on a semilogarithmic scale.

reasonably well with the result reported by Blagoev et al. [8], but the heavily cascaded nature of the decay curves gave us low confidence in the validity of multiexponential fitting for these decays. For the $^2P_{3/2}$ decay curves, the parameters obtained from a two-exponential fit were substantially shorter than the value reported by Blagoev et al. [8]. The second phase of the analysis involved the use of the ANDC method of joint analysis of cascade correlated decay curves, using computer codes which employ both a direct [9] and a noise filtered [23] approach.

For the $7p\,^2P_{1/2}$ level, our analysis indicated that cascades from both the 8s and 7d levels were required for a consistent joint analysis. Although transitions from higher lying ns and nd levels were not apparent in our spectra, we checked their possible influence on our ANDC analysis by constructing simulated decay curves using the lifetimes for $n \leqslant 10$ computed by the Coulomb approximation [19]. The correlated analysis rejected these decay curves by as-

signing them insignificantly small normalizing factors. It is interesting that an ANDC analysis for the 6p levels in Hg II by Pinnington et al. [24] indicated that there the cascading from 6d dominates, with the contribution from 7s being negligible. This is not surprising, since the 6p decay is not affected by cancellation, and it is repopulated along the yrast chain. Since the ANDC method provides a set of independent relationships among the decay curves for each distance downstream, distortions in the decay curves due to escape from the viewing volume would be revealed by variations in the extracted lifetime as a function of distance downstream. Our ANDC measurements yielded a lifetime of 14.5 ± 1.0 ns for the $7p\,^2P_{1/2}$ level, significantly shorter than the simple curve-fitted result. For the $7p\,^2P_{3/2}$ the long-lived cascades played a much smaller role in the ANDC analysis, and the best results were obtained including only the repopulation from 7d. Here the results of the ANDC and the curve fitting analyses were in basic agreement, and yielded a lifetime of 1.2 ± 0.2 ns. These results are compared to the various theoretical computations in table 1.

One- and two-exponential fits were made to the 6p–8s and 6p–7d decay curves, which yielded lifetimes $\tau(8s\,^2S_{1/2}) = 2.6 \pm 0.3$ ns and $\tau(7d\,^2D_{3/2}) = 3.0 \pm 0.3$ ns. The $7d\,^2D_{5/2}$ decay curve was blended with an Hg III line, and was not used in the analysis. Our measured 8s lifetime was significantly shorter than the value 5.5 ns obtained using the Coulomb approximation [19], and is somewhat shorter than the value 3.9 ns predicted by Migdalek [25]. Our measured $7d\,^2D_{3/2}$ lifetime is in good agreement with the value 2.7 ns obtained using the Coulomb approximation [19], but is substantially shorter than the value 5.0 ± 0.6 ns measured by Blagoev et al. [8].

In conclusion, our results verify that the Hg II $7p\,^2P_{1/2}$ and $^2P_{3/2}$ differ by more than a factor of ten in lifetime, and provide accurate values for both lifetimes that are substantially shorter than the results reported by Blagoev et al. [8]. This confirms the assertion that the decays of these levels are strongly affected both by cancellation and by configuration interaction, and indicates the difficulties that such situations pose to theoretical predictions. Furthermore, the 923 Å line provides an accurately measured example of a low-lying resonance transition, long of the Lyman limit, which can be strong in emission because of heavy cascading, but weak and unsaturated in absorption because of its unusually long lifetime.

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