Lifetimes of $5d^9 6p$ and $5d^8 6s\,6p$ levels in Hg III

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Experimental and theoretical studies of lifetimes and transition probabilities for doubly ionized mercury, Hg III, are reported. Such data are currently of great astrophysical interest because observations with the Goddard high resolution spectrograph on board the Hubble space telescope have shown some $5d^9 6s$-$5d^9 6p$ transitions of Hg III in the spectrum of the chemically peculiar star χ Lupi. We now report experimental lifetimes for some selected $5d^9 6p$ and $5d^8 6s\,6p$ levels, determined by beam-foil spectroscopy, and compare the results with theoretical transition probabilities based on relativistic multiconfiguration Dirac-Fock calculations.

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I. INTRODUCTION

One of the first projects on the Hubble space telescope (HST) was the registration of a high-resolution spectrum of a chemically peculiar star, χ Lupi, which has a high abundance of mercury. The analysis of one spectrum [1] verified the earlier finding of an isotope anomaly of Hg in the star's atmosphere. Mercury is here present in the form of ^{204}Hg to 99%, whereas this isotope is only a minor contributor (7%) in the solar system. The study of Leckrone, Wahlgren, and Johansson [1] was based on the resonance line of singly ionized mercury, Hg II, at 1942 Å.

A possible explanation for this isotope anomaly in χ Lupi is a delicate balance between the radiative pressure and gravitation. This is the major ingredient in the diffusion model developed by Michaud [2]. Determinations of the abundance and isotopic composition of neutral and doubly ionized mercury would thus constitute a critical test of this model, and observations of the spectral regions containing Hg I and Hg III lines have therefore been performed with the HST. The signal-to-noise ratio and the resolution of the spectra permit accuracies to within a few percent in the determination of isotopic compositions, provided that the relevant atomic data (energy levels, wavelengths, oscillator strengths, etc.) are available. This is the case for Hg I and Hg II, whereas the information is more fragmentary for Hg III, particularly concerning transition probabilities.

Because the wavelength interval accessible with the HST at the highest spectral resolution has a lower limit of about 1700 Å [the side-B detector of the Goddard high resolution spectrograph (GHRS) is out of function], the observations are restricted to lines above 1700 Å. The relevant Hg III lines observed with the HST appear at around 1740 Å.

In Hg III, which is isoelectronic to Pt I and Au II, the ground term is $5d^{10}\,{}^1S_0$. The lowest excited even configurations $5d^9 6s$ and $5d^8 6s^2$ and the odd configurations $5d^9 6p$ and $5d^8 6s\,6p$ were established many years ago [3]. Recently Wyart *et al.* [4] have also provided energies of levels belonging to the $5d^9 6d$ and $5d^9 7s$ configurations.

With the single exception of the electron-excitation experiment of Blagoev and Dimitrov [5] (who determined the lifetimes of some $5d^8 6s^2$ levels) no experimental or theoretical lifetimes or f values have been reported for Hg III. The present study was undertaken to provide such information for transitions from the $5d^9 6p$ levels. The $5d^9 6p$ configuration consists of 12 levels with J values ranging from 0 to 4. These levels decay to the $5d^{10}\,{}^1S_0$ ground term and the four $5d^9 6s$ levels. Using the beam-foil excitation method we have now measured lifetimes for three $5d^9 6p$ levels and one $5d^8 6s\,6p$ level. This work was combined with theoretical investigations, utilizing the multiconfiguration Dirac-Fock (MCDF) package GRASP [6] and the program suite of Cowan [7]. The calculations will be only briefly discussed in this paper, while a comprehensive report is planned to be published separately [8].

The lifetimes obtained in beam-foil experiments tend to be of lower precision than those based on various laser excitation methods, cf. the study of Hg I by Benck, Lawler, and Dakin [9]. However, the laser techniques are very difficult to apply to doubly ionized species, in particular when the transitions are in the vacuum ultraviolet and the lifetimes are very short (2 ns or less). This holds for the $5d^9 6p$ levels of Hg III for which theory [8] predicts lifetimes in the 0.3–1-ns range. In cases like this, the beam-foil technique is the only realistic alternative.

II. EXPERIMENT

Ions of $^{202}Hg^+$ were accelerated to an energy of 250 keV by the University of Toledo heavy ion accelerator (produced by Danfysik, Jyllinge Denmark) and sent through a thin, self-supporting carbon foil. To minimize energy loss, straggling, and multiple scattering (the last named of which can lead to a divergent beam on the downstream side), very thin foils (2.0–2.4 $\mu g/cm^2$, with 50% uncertainties) were used. The radiation emitted by the foil-excited ions was dispersed with an Acton 1-m normal incidence vacuum monochromator. Survey spectra were taken in the region 1900–3000 Å, using a cooled Centronic detector. In the region 1200–1900 Å, expected to be of particular interest to the HST observations, a solar-blind Hamamatsu detector was used, whereas light between 700 and 1200 Å was detected with a Channeltron. The lifetimes were obtained by measuring the intensity of the spectral lines as a function of the distance along the beam from the foil. The recorded decay curves were normalized to the intensity of undispersed light, measured with a photomultiplier viewing the beam via a fiber optical link about 5 mm downstream from the foil. In this way it was possible to compensate for variations in the beam intensity or possible changes of the foil properties during a measurement. The accelerator and the experimental setup have been described in detail by Haar [10] and shorter summaries are also available [11,12].

Beam-foil experiments using heavy ions of relatively low energy present a number of specific problems which require detailed analyses. A troublesome technical limitation worthy of mention is the shortness of the typical foil failure time. In the present experiment with a beam intensity of 4–8 nA/mm^2 the failure time was typically 4–5 min, which placed a practical upper limit on the time available for recording a particular decay curve. Since replacing a foil in the midst of such a measurement introduces new uncertainties, a larger number of decay curves of moderate statistical accuracy were necessarily taken instead of fewer higher-quality ones, the usual way in lifetime studies using the beam-foil technique. The problems of energy loss, energy straggling, and scattering in the foils have been discussed by several authors, for instance, [13,14].

The energy of the ions before entering the foil was determined from the terminal voltage of the accelerator, which was calibrated to better than 1% by the use of quantum beat measurements in neutral helium. The velocity of the Hg ions after transmission of the foil was determined using the following considerations. The energy loss of the ions in the foil was specified using the computer program TRIM, which is described in Ref. [15]. For 250-keV ions in carbon this gives the electronic stopping $-(dE/dx)_e = 6.4$ keV $cm^2/\mu g$ and the nuclear stopping $-(dE/dx)_n = 15.8$ keV $cm^2/\mu g$. Several authors, e.g., Hvelplund *et al.* [16] and Garnir-Monjoie and Garnir [17], have shown that only a fraction of the nuclear stopping (typically 30%) should be included for ions emerging in the forward direction, i.e., the ions normally studied in beam-foil experiments. With 2-$\mu g/cm^2$ foils this gives an average energy loss of 23 ± 10 keV, where our uncertainty estimate has been made on the conservative side. We also estimated the angular distribution of the ions after the foil, using the formalism developed by Sigmund and Winterbon [18]. For a Hg beam of 250 keV passing through a carbon foil with thickness 2 $\mu g/cm^2$, the half-width of the angular distribution is predicted to be about 3°. These effects were all taken into account when estimating the uncertainty of the final results.

As in most beam-foil measurements, cascade repopulation should be taken into account. In the present case, the $5d^96p$ levels are expected to be predominantly fed from $5d^96d$ and $5d^97s$. The ideal way of correcting for such cascading would have been to use the so-called arbitrarily normalized decay curve (ANDC) technique [19], i.e., to jointly analyze the primary decays (of the $6p$ levels) and the decays of the radiation from the $6d$ and $7s$ levels, specified by the wavelength data given by Wyart *et al.* [4]. However, this was precluded because only a few weak $6p$-$6d$ and $6p$-$7s$ lines appeared in our spectra. This is not surprising, because of the relatively low beam energy, which does not favor the production of doubly charged Hg when passing the foil. Thus measurements by Heinemeier *et al.* [20] imply that for 250-keV Hg^+ ions, only 11% are doubly charged after the foil (40% are neutral and 49% are singly charged). This fact, consistent with our more qualitative data on spectral-line intensities, necessitated the use of the less rigorous curve-fitting method for lifetime determinations. Each of the measured decay curves was fitted to one or two exponential forms. However, while this method leads to larger uncertainties in the final lifetimes, experience has shown that cascading is not the dominant source of error in lifetime measurements using heavy, slow ions. In many such cases beam-foil data have subsequently been corroborated by more precise laser measurements. A very interesting result was reported by Andersen *et al.* [21], who found that in these systems the beam-foil excitation predominantly tends to populate low-lying states. A model proposed by Kemmler, Burgdörfer, and Reinhold [22] suggests that the mechanism that leads to the unusually large populations of high n,l states in beam-foil excitation of lighter ions requires collisional frequencies that exceed orbital frequencies, a condition not favored for slower sub-MeV heavy ions.

Because of the relatively small fraction of Hg^{2+} after the foil we had to use a comparatively wide monochromator entrance slit, obtaining typical linewidths of 1.0–1.5 Å below 1200 Å and 2–3 Å above 1200 Å. In the region 1200–1900 Å this led to considerable blending of lines (see below) and effectively precluded decay measurements for the $6s$-$6p$ transitions. However, three of the 12 levels of the $5d^96p$ configuration have $J=1$, and these can decay to the $5d^{10}\,{}^1S_0$ ground state with allowed transitions in the wavelength range 740–850 Å, a region where the density of lines is lower. We were therefore able to determine the lifetimes of these three levels as well as for one $5d^86s6p$ level with $J=3$ which has a decay channel in this region. To test the measurements and the reliability of the various corrections, we also measured the decay times of some strong Hg I and Hg II transitions, for which reliable experimental and theoretical data are

available, see, e.g., Pinnington *et al.* [23]. The following lifetime values were thereby obtained: 1.49 ns (Hg I, $6s6p\ ^1P$), 2.1 ns (Hg II, $6p\ ^2P_{3/2}$), and 1.7 ns (Hg II, $6d\ ^2D_{5/2}$). Although less accurate, these results are in satisfactory agreement with most of the recent results.

III. THEORETICAL CALCULATIONS

Two types of calculations were carried out to obtain theoretical transition probabilities and lifetimes. The primary method employed the relativistic multiconfiguration Dirac-Fock code GRASP [6] with extensive inclusion of configuration interaction. Corroborative calculations with fewer configurations were also performed using the program suite of Cowan [7], in which relativistic corrections to the Schrödinger formulation are made, with the Slater integrals adjusted so as to fit the experimental energy levels. The GRASP calculations are carried out in several steps. First the MCDF equations are solved to obtain the large and small component radial functions which, together with the spinor spherical harmonics, form the relativistic one-particle orbitals. Antisymmetrized sums of products of these orbitals are used to obtain the configuration state functions (CSF's). These functions are then used to set up a configuration-interaction (CI) calculation, using a large set of formed CSF's by creating and diagonalizing the Hamiltonian matrix. Here the low-frequency limit of the Breit interaction is also included. Each J value and parity were optimized individually, either on the basis of the weighted average energy of all CSF's (AL), or on one level (OL), or a set of levels (EOL). The line strengths are then computed using the Babushkin form of the transition operator and the eigenvectors obtained in the CI calculation. The transition probabilities are obtained using both experimental and theoretical wavelengths, with the standard $1/\lambda^3$ dependence. The differences illustrate the magnitude of the inaccuracies in the theoretical energy differences. The transition probabilities were summed over final states to obtain lifetimes. A more comprehensive report of these calculations is planned to be published elsewhere [8].

IV. DATA ANALYSIS AND RESULTS

A. Spectra

As a preparation to the lifetime determinations, the beam-foil spectra were studied in detail. In the region 1200–1900 Å a number of Hg III $6s$-$6p$ lines appeared, but they were frequently blended by stronger Hg II or Hg I multiplets or even masked by intense C I–C III transitions, the latter originating from excited species sputtered from the foil. In the region of primary HST interest we identified a peak at 1738.5±0.5 Å in our beam-foil spectra, but its width of about 3 Å [full width at half maximum (FWHM)] did not allow it to be resolved into the three expected Hg III lines with tabulated wavelengths of 1738.48, 1738.53, and 1740.27 Å, and this fact ruled out a lifetime measurement. However, to test the theoretical predictions for Hg III (Joueizadeh *et al.* [8]) other transitions in this spectrum can be selected for

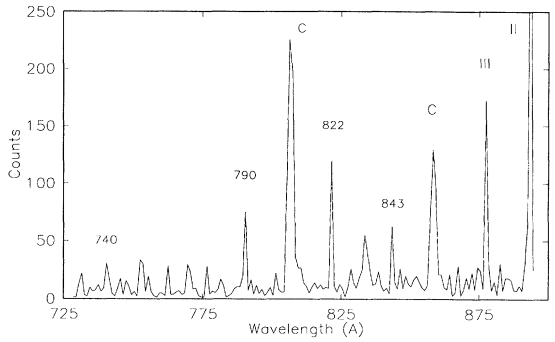

FIG. 1. Survey spectrum of Hg in the region 725–900 Å. The Hg III lines for which decay times were measured are indicated by wavelength. For a more detailed discussion of the spectrum, see text.

study. Since no Hg I lines and only a few Hg II lines are expected below 900 Å, whereas several Hg III transitions have wavelengths in the 700–900-Å interval, we decided to concentrate on this region.

A partial spectrum is shown in Fig. 1. Three lines, at 740.75, 790.17, and 843.11 Å, can be identified as transitions from the three $J=1$ levels of the $5d^9 6p$ configuration to the $5d^{10}\ ^1S_0$ ground state, whereas a fourth line, at 822.69 Å, is a $5d^9 6s$-$5d^8 6s6p$ transition. These lines were selected for lifetime measurements. More than 15 of the other lines in Fig. 1 also belong to Hg III, but they were either blended (this being the case for the peak labeled III in Fig. 1 which consists of two $5d^9 6s$-$5d^8 6s6p$ lines, at 878.59 and 879.29 Å [3]) or too weak to allow lifetime measurements. Note also the intense Hg II line at 893.11 Å ($6s\ ^2S_{1/2}$–$7p\ ^2P_{3/2}$) and the relatively strong C II lines close to 807 and 858 Å. The larger widths of the latter reflect the low kinetic energies of the sputtered C ions. (The monochromator was refocused so as to produce narrow lines for the radiation emitted by the much faster Hg ions.)

B. Lifetimes

The decay curves of two of the $6p$ levels with $J=1$ are shown in Figs. 2 and 3. For each of the Hg III lines mea-

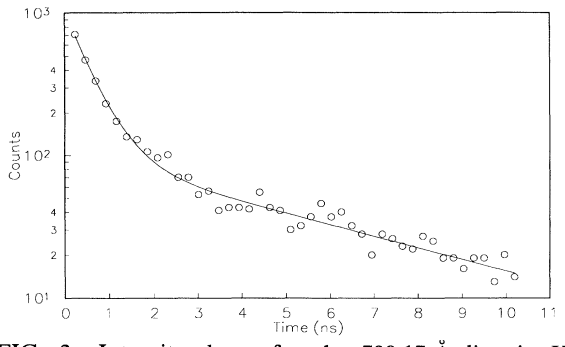

FIG. 2. Intensity decay for the 790.17-Å line in Hg III, $5d^{10}\ ^1S_0$-$5d^9 6p\ ^1P_1$. The solid line denotes the two-exponential fit.

TABLE I. Lifetimes of excited levels in Hg III.

Transition[a]	Energy (cm^{-1}) of upper level[b]	Wavelength (Å)	Lifetime (ns) Experiment	Theory[c]
$5d^{10}\,{}^1S_0-5d^96p\,{}^3D_1$ $5d^9_{3/2}6p_{3/2}$ $J=1$	134 998	740.75	0.90 (15)	0.70; 0.63
$5d^{10}\,{}^1S_0-5d^96p\,{}^1P_1$ $5d^9_{5/2}6p_{3/2}$ $J=1$	126 556	790.17	0.52 (15)	0.28; 0.26
$5d^{10}\,{}^1S_0-5d^96p\,{}^3P_1$ $5d^9_{3/2}6p_{3/2}$ $J=1$	118 607	843.11	1.20 (15)	1.00; 0.88
$5d^96s\,{}^3D_2-5d^86s6p$ $J=3$	167 580	822.69	0.55 (15)	

[a]For the three $5d^96p$ levels both nominal *LS* and *jj* labelings are given.
[b]Moore, Ref. [3].
[c]Results obtained from calculations with the GRASP code (see text). In each case the first value has been calculated using theoretical transition wavelengths and the second one using experimental wavelengths.

sured, four decay curves were selected for a detailed analysis. After the subtraction of a small background the curves were fitted to one or two exponential forms, using the program DECCEL, written by Haar [10]. It can be noted that the decay curves of the 790.17- and 843.11-Å lines (Figs. 2 and 3) require a two-exponential fit. The final lifetime values are given in Table I where we also compare them with our theoretical results based on the GRASP code. The states involved are in intermediate coupling but we shall designate them by nominal *LS* and *jj* labels.

The experimental uncertainties given in Table I were estimated as follows. The statistical uncertainties of the data are typically 4–6 %. We estimate the velocity uncertainty after the foil to be about 3%. Since cascading from higher levels has been taken into account in an approximate way (multiexponential fitting) instead of the more rigorous ANDC technique, this introduces another uncertainty in the results. A conservative estimate, largely based on our experience of similar beam-foil studies for other complex ions, would here be 10%. By quadrature addition we arrive at the uncertainties given in Table I.

Table I clearly shows that the experimental lifetimes are somewhat longer than the theoretical ones. In particular, a fairly significant difference can be noted for the $5d^96p\,{}^1P_1$ level (790.17 Å). In cases where substantial cascade repopulation is present, multiexponential fitting methods can overestimate lifetimes. However, such cascading was not evident either in the corresponding transition intensities or as secondary exponentials in the fitted decay curves. We also considered the possibility of overlooking a prompt initial component on the decay curves (corresponding to a lifetime of 0.30 ns or less). While no direct evidence of this could be found, this possibility cannot be ruled out entirely. Furthermore, the 790.17-Å line is bracketed by Hg III lines, at 788.53 and 792.31 Å [3]. The intensity of each of these lines is about 25% of that of the 790.17-Å transition in our spectra, and although the three lines were practically resolved some interference cannot be ruled out.

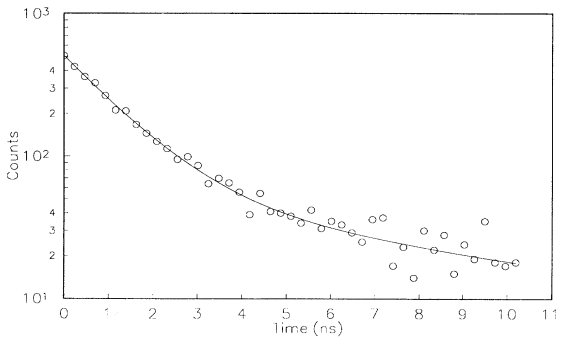

FIG. 3. Intensity decay for the 843.11-Å line in Hg III, $5d^{10}\,{}^1S_0$-$5d^96p\,{}^3P_1$. The solid line denotes the two-exponential fit.

Table I also includes the experimental result for the 822.69-Å line which is a transition between levels of two excited configurations, $5d^96s$ and $5d^86s6p$. Although $5d^86s6p$ was included in the optimization of the astrophysically important singly excited states, the doubly excited states were not themselves optimized. Thus no theoretical comparison is given for this transition.

The three $5d^96p$ ($J=1$) levels combine not only with the $5d^{10}$ ground state but also with three of the four $5d^96s$ levels. Transition probabilities for these channels, calculated with the GRASP code, are given in Table II. Of these the transition probabilities between $5d^{10}$ (closed $5d$ subshell) and $5d^96p$ (open $5d$ subshell) are the least accurate in the calculations, because of the large difference in the $5d$ orbital in the two configurations. The transition probabilities between excited $5d^96s$ and $5d^96p$ states are much easier to calculate and the values in Table II should be quite reliable. They agree to within 10–20 % with the results of our less elaborate calculations, based on the Cowan programs [7].

V. CONCLUDING REMARKS

We have reported experimental lifetimes for some $5d^96p$ and $5d^86s6p$ levels in Hg III, and corroborated the data with two types of theoretical calculations. Considering the approximations needed in theoretically treating this complex many-electron system and the experimental difficulties discussed above, the agreement between theory and experiment must be considered as quite satis-

TABLE II. Probabilities (in s^{-1}) for transitions and lifetimes (in ns) for $5d^9 6p\ J=1$ states in Hg III. $X[n]$ implies $X \times 10^n$.

	Upper level (energy)		
Lower level	$5d^9_{3/2}6p_{1/2}$ (118 607 cm^{-1})	$5d^9_{5/2}6p_{3/2}$ (126 556 cm^{-1})	$5d^9_{3/2}6p_{3/2}$ (134 998 cm^{-1})
$5d^{10}\ J=0$	2.30[8]	3.03[9]	5.14[8]
$5d^9 6s\ J=2$	8.30[8]	1.76[8]	3.87[7]
$5d^9 6s\ J=1$	1.74[6]	8.14[7]	9.31[8]
$5d^9 6s\ J=0$	7.09[7]	5.21[8]	1.03[8]
Lifetime	0.88	0.26	0.63

factory.

From the experimental results (lifetimes) only sums of transition probabilities (gf values) can be directly deduced. Unless branching ratios are known, it is not possible to obtain information about individual gf values. However, a comparison of experimental and theoretical lifetimes still yields valuable insight into the accuracy of theoretical calculations, thereby providing indirect information as to gf values. One way of obtaining branching ratios would be to use the theoretical values for the transitions between excited states ($5d^9 6s$-$5d^9 6p$) and deduce the transition probabilities for the $5d^{10}$-$5d^9 6p$ resonance lines with the help of experimental lifetimes. In summary, although a certain difference persists between the experimental and theoretical lifetimes, we can conclude that the theoretical values are accurate to within 15% which should be quite sufficient in the quantitative analyses of HST spectra of peculiar stars.

For a more detailed work on Hg III, ion beams of significantly higher energy than that used by us (250 keV) would be needed. According to Heinemeier *et al.* [20] the optimal energy for producing Hg^{2+} is close to 1 MeV. At this energy higher spectral resolution should be possible, and the precision in lifetime measurements could be improved because reduced angular scattering would yield better definition of the time scale after the foil. On the other hand, the presence of higher charge states, notably Hg^{3+} and Hg^{4+}, would greatly complicate the analyses. Thus the spectra Hg IV and Hg V, which have been only partially analyzed by Joshi, Raassen, and Arcimowicz [24] and Raassen, Jashi, and Tauheed [25], respectively, are very line rich. This fact could result in blending and masking of the Hg III lines now studied. Finally, to corroborate the Hg III data, we have now initiated similar experimental and theoretical studies of Au II (isoelectronic to Hg III), the transitions of which have also been identified in stellar spectra from the Hubble space telescope.

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