Lifetimes of the $5d^9 6p$ levels in Au II

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Received November 4, 1992; revised manuscript received February 1, 1993

We report experimental lifetimes for 11 of the 12 $5d^9 6p$ levels of Au II, determined by means of the beam-foil excitation method. The data are compared with theoretical calculations based on the Cowan group of programs. These first studies of Au II lifetimes and transition probabilities were motivated largely by the need for such data in the analysis of high-resolution stellar spectroscopy done with the Goddard High-Resolution Spectrograph on the Hubble Space Telescope.

1. INTRODUCTION

In a recent paper[1] we reported lifetime measurements for levels in doubly ionized mercury, Hg III, performed by the beam-foil excitation technique. This research was partly motivated by the observation of Hg III transitions in high-resolution spectra of the chemically peculiar B star χ Lupi[2] obtained with the Goddard High-Resolution Spectrograph (GHRS) on the Hubble Space Telescope (HST). In the echelle mode of the GHRS, a resolving power of approximately 100,000 can be achieved, but only one echelle order (~10 Å) can be monitored at a time. The region around 1740 Å, where some Hg III lines appear, also contains the strongest $6s$–$6p$ transition ($5d^9 6s\ ^3D_3$–$5d^9 6p\ ^3F_4$ in LS notation) in Au II, isoelectronic to Hg III. The wavelength of this transition was recently determined to be 1740.476 Å by means of the vacuum-ultraviolet Fourier-transform spectrometer at the University of Lund. This value agrees with the wavelength measured on the HST to within 2–3 mÅ and represents an improvement by a factor of approximately 20 over the tabulated value[3] of 1740.52 Å. This line is prominent in the GHRS spectrum of χ Lupi.[4]

Analyses of stellar data, performed with recent unpublished calculations of gf values for Au II, yield for this star an overabundance of gold (compared with the solar value) by approximately 4 orders of magnitude. This is comparable with the abundance pattern found in mercury and platinum.[4] Various explanations of such high abundances are now being investigated by means of systematic studies of the heavy elements. In such work accurate experimental data (level energies, wavelengths, oscillator strengths, lifetimes, hyperfine-structure separations, isotope shifts, etc.) are required. In the present paper we report results of experimental and theoretical studies of lifetimes for Au II, which is isoelectronic to Pt I and Hg III.

The ground state of Au II is $5d^{10}\ ^1S_0$, and the two lowest even configurations are $5d^9 6s$ and $5d^8 6s^2$. The lowest odd configuration is $5d^9 6p$ (Fig. 1). For Au II the present knowledge of energy levels can be considered satisfactory (although the important $5d^8 6s 6p$ configuration is practically unknown), whereas no data on lifetimes or oscillator strengths have appeared in the literature.

2. EXPERIMENT

The Danfysik heavy-ion accelerator at the University of Toledo was used to accelerate ions of $^{197}Au^+$ to 220 keV. After mass analysis, the ions were directed through a thin self-supporting foil of carbon. The ions were produced from metallic gold heated in the high-temperature oven of a Danfysik Model 911A ion source. Foil thicknesses were 2.0–2.4 $\mu g/cm^2$. The light emitted by the foil-excited ions was analyzed with an Acton 1-m normal incidence vacuum monochromator, equipped with a 1200-lines/mm concave grating, blazed at 1500 Å. The photons were counted at the exit slit with a solar-blind Hamamatsu detector (1200–1900 Å) or with a dry-ice-cooled Centronic detector (1900–2500 Å). We determined the atomic lifetimes by recording the intensity of the spectral lines as a function of the distance from the foil. To compensate for fluctuations of the ion-beam current, an optical normalization technique was used. For details about the experimental setup see Refs. 5 and 6.

In beam-foil experiments with heavy ions of comparatively low kinetic energy the energy loss in the foil amounts to a significant fraction that must be accurately known and taken into account when lifetime data are analyzed. The energy loss can be divided into two parts, which are often called the electronic and the nuclear stopping fractions. We computed these fractions with the computer program TRIM, developed by Ziegler *et al.*[7] For 220 keV ions of $^{197}Au^+$ penetrating a carbon foil, the electronic energy loss amounts to 6.0 keV $cm^2/\mu g$ and the nuclear loss to 15.5 keV $cm^2/\mu g$. Only a fraction of the latter, typically 30%, should be taken into account for foil-excited ions moving in the forward direction, which is the usual case in beam-foil experiments.[8,9] In the present case (with 2-$\mu g/cm^2$ foils) we thus assume that the total loss amounts to 21 ± 10 keV. Foil-thickening effects were estimated,[10,11] but for our experimental conditions these effects are within the quoted tolerances of the foil thicknesses. The energy of the incoming ions is known with a relative uncertainty of ~1% (through calibration with quantum-beat measurements), and we can therefore assume that the velocity after the foil is 0.443 ±

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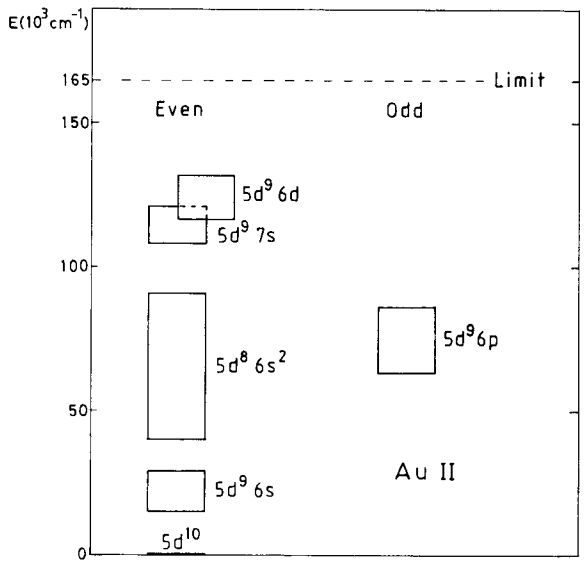

Fig. 1. Schematic energy-level diagram of Au II, showing some low-lying configurations.

0.013 mm/ns, where the error estimate should be on the conservative side. Because of multiple scattering in the foil, the ion beam after the foil is no longer parallel but is slightly divergent. We used the tables of Sigmund and Winterbon[12] and found that the angular distribution of the ions has a half-width of ~3°. This effect was also taken into account in the analysis of the data.

One practical problem encountered in beam-foil studies that use heavy ions of low energy involves the shortness of foil failure times. In the present work these times were ~4–5 minutes. This requires rapid positional scanning, since the replacement of a foil in the midst of the recording of a decay curve would introduce systematic uncertainties. Mesh-mounted foils can extend failure times for wavelength measurements, but for lifetime studies their gradual facet-by-facet failure leads to difficulties in normalization by either optical or beam current monitoring. To achieve the desired statistical accuracy it was necessary to record a large number of rapidly scanned decay curves of moderate statistical accuracy instead of a smaller number with high statistical accuracy.

The beam-foil excitation is far from selective, implying that many levels in a given ion can be populated. This results in a feeding of the levels under study from higher states (cascading), thereby complicating the analyses of decay curves. Solutions have been found to this problem, notably the arbitrarily normalized decay curves (ANDC) method of Curtis *et al.*,[13] which is based on joint analyses of the decay curve of the level under study and of the curves of the levels predominantly feeding that level. However, while this technique of analysis works well in simpler systems, where repopulation is dominated by a small number of strong cascades, its applicability is limited in complex spectra, where the level under study is fed by a large number of weak cascades. Au II appears to have such a complex spectrum, and the situation is further complicated by the fact that the energies of the cascading levels are only fragmentarily known. Thus we were forced to fit the decay curves to one or two exponential forms. While this method must be used with great care, several studies indicate that this approach gives satisfactory results for low charge states of beam-foil excited heavy atoms. In such systems curve-fitted beam-foil results have frequently been confirmed by subsequent laser experiments, in which the excitation is selective and no cascading thus occurs. For example, in Si I the beam-foil measurements by Bashkin *et al.*[14] were confirmed by the laser measurements by O'Brian and Lawler,[15] and in Mn II the beam-foil measurements by Martinson *et al.*[16] were confirmed by the laser measurements by Pinnington *et al.*[17]

3. DATA ANALYSES AND RESULTS

A. Spectra

In the wavelength region studied (1200–2500 Å), approximately 80 lines could be assigned to transitions in Au I–Au III by means of the available spectroscopic literature on these spectra.[18–22] In general, lines in Au I and Au II were more intense than those belonging to Au III, which is consistent with measurements of the post-foil charge distributions of heavy ions. While no calculated data[23] for Au ions in the 200–250-keV range have been reported, measurements have been carried out for Hg^+ ions.[24] For 220-keV Hg^+ ions incident upon carbon foils, the charge distribution is 37% (0), 52% (+1), and 11% (+2). We assume the distribution for Au to be quite similar. This assumption is consistent with our spectral data, which show the presence of some Au III lines, but these are fairly faint when compared with the lines of Au I and Au II. This fact is important in the discussion of possible blends, as discussed below.

A spectrum is shown in Fig. 2. Note that the Au II line at 1740 Å (which is important for the HST data) is particularly intense. For most of the other Au II transitions indicated in Fig. 2, decay times were determined in the present work.

Before we attempted to measure lifetimes, we tried to identify as many $5d^9 6s$–$5d^9 6p$ lines as possible in the spectra. Decays of 11 of the 12 $5d^9 6p$ levels were established, and only the $6p$ level ($J = 0$) escaped observation in the present work. Thus no line at 1823.23 Å [which, according to Platt and Sawyer,[19] is the $6s$ ($J = 1$)–$6p$ ($J = 0$) combination] appeared in the beam-foil spectra. Interestingly, this line has not been listed in the more recent analysis by Ehrhardt and Davis,[20] which may indicate that the early identification[19] should be revised.

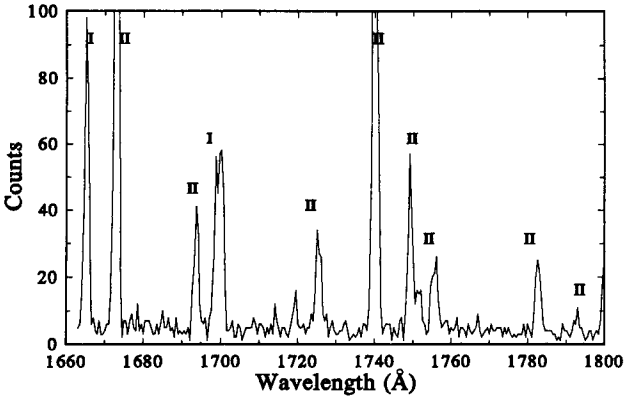

Fig. 2. Partial beam-foil spectrum of Au. The ionization stages of the strongest lines are indicated by Roman numerals.

Table 1. Lifetimes of the $5d^9 6p$ Levels in Au II

Level	J	Energy (cm^{-1})	Wavelength (Å)	Lifetime (ns) Experiment	Theory
1°	2	63053	2082.1	3.8 ± 0.5	2.5
2°	3	65004	2000.8	3.5 ± 0.5	2.2
			2110.7		
3°	4	72495	1740.4	2.4 ± 0.3	1.4
4°	2	73178	1800.6[a]	2.6 ± 0.5	1.6
			1720.0		
5°	1	73404	1793.3[a]	2.3 ± 0.4	1.5
6°	3	74792	1749.7	2.4 ± 0.4	1.4
7°	2	76660	1694.3[a]	2.5 ± 0.4	2.2
			2125.3		
8°	1	81660	1224.6[b]	1.4 ± 0.3	0.5
10°	3	85700	1783.2	2.5 ± 0.4	1.6
11°	1	85708	1725.8[a]	1.8 ± 0.3	0.7
12°	2	86566	1756.1[a]	2.4 ± 0.4	1.6

[a]Line possibly blended by a Au III transition.
[b]Decay curve partly distorted by Ly-α (1216 Å).

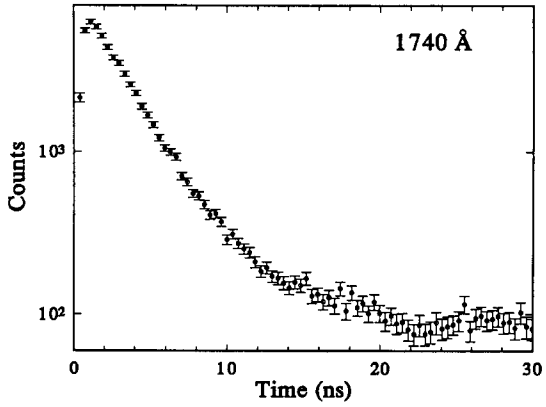

Fig. 3. Decay curve of the Au II transition at 1740 Å (decay of the $5d^9 6p, J = 4$ level).

B. Lifetimes

The lifetimes of 11 of the 12 levels of the $5d^9 6p$ configuration were measured, and the results are presented in Table 1. A characteristic decay curve for the 1740-Å line is shown in Fig. 3. Before we discuss our results in detail, we should note that we tested our setup and our data analysis methods by measuring the lifetime of the $6p\ ^2P_{3/2}$ level in Au I, for which reliable data were previously available. From decay measurements of the 2428-Å resonance line ($6s\ ^2S_{1/2}$–$6p\ ^2P_{3/2}$), a lifetime of 4.5 ± 0.5 ns was obtained, in good agreement with a previous beam-foil result,[25] 4.3 ± 0.5 ns, and a more accurate value, 4.6 ± 0.2 ns, based on a laser excitation study.[26]

The error limits in Table 1 are based on the following considerations. The statistical uncertainties in the data are typically 4–8%, whereas the velocity after the foil is known to within 3%, as noted above. The uncertainty introduced when one corrects for cascading by means of multiexponential fits instead of the ANDC method can be estimated to 6–8%. We use quadrature addition to arrive at the uncertainties given in Table 1. However, in a few cases some systematic errors from line blending cannot be entirely ruled out. These cases are indicated in Table 1 and will be discussed below.

The decay of the 4° level (73178 $cm^{-1}, J = 2$) was measured from two lines, 1800.6 and 1720.0 Å. The former may be partially blended by an Au III transition at 1802.0 Å in our spectra, but this blending appears to be less serious because the decay times for the 1800.6- and 1720.0-Å lines were practically identical. Similarly, the 1793.3-Å line (5°, 73404 $cm^{-1}, J = 1$) could not be spectrally resolved from an Au III line at 1793.76 Å. The intensity of the latter line has been estimated to be 500. On the other hand, an Au III line at 1775.2 Å, intensity 800, was barely visible in our spectra, thereby supporting the lifetime obtained for the Au II level. The most serious blends probably concern the 7° (76660 $cm^{-1}, J = 2$) and 11° (85708 $cm^{-1}, J = 1$) levels. In the former case the Au II line at 1694.3 Å is close in wavelength to the 1693.9-Å transition in Au III (intensity 1000), but fortunately the lifetime of the 7° level could also be determined from the nearly unblended Au II line at 2125.3 Å, and the results for the two different lines were consistent to within 10–15%. In the case of the 11° level, the Au II line at 1725.8 Å could be only partly separated from an Au III transition at 1727.3 Å (intensity 500). Similarly, the Au II line at 1756.1 Å (12°, 86566 $cm^{-1}, J = 2$) is close to the Au III line at 1756.9 Å (intensity 500). However, another Au III line (1746.1 Å, intensity 500) failed to show up in our spectra, and this fact increases the confidence in our lifetimes for the 11° and 12° levels of Au II. Finally, the decay curves of the 1224.6-Å line (8°, 81660 $cm^{-1}, J = 1$) all displayed an unusual shape. After an initial exponential decay (by a factor of ~20) during approximately the first 10 ns after the excitation, the intensity began to increase again. This could be explained by Doppler-shifted 1216-Å H I Ly-α radiation (from hydrogen sputtered from the foil or from possible hydrocarbon buildup on the foil surface), which was reflected into the monochromator when a certain region of the foil-excited beam was viewed. Although the 1224.6-Å and Ly-α lines are well separated in wavelength, the latter was approximately 20 times more intense than the former, close to the foil. Thus the first part of the decay curves could still be used for the analysis. The uncertainty estimate of the lifetime of the 8° level has been increased to take into account the truncation of imperfect data.

A study of Table 1 reveals some interesting lifetime systematics for the $5d^9 6p$ levels under study. The lifetimes of the two lowest levels are markedly longer than those of the remaining levels. This fact is not unexpected because of the significantly longer wavelengths involved in the decay of the 1° and 2° levels. The lifetime values of the higher levels fall within 10% (2.4–2.6 ns), with the exception of the levels with $J = 1$. Here the explanation is that these three levels (5°, 8°, and 11°), in addition to decaying to $5d^9 6s$ ($J = 1$ and $J = 2$), also make allowed transitions to the $5d^{10}\ ^1S_0$ ground term. From the measured lifetimes, estimates can be made of the transition probabilities of these resonance lines.

4. THEORETICAL CALCULATIONS

We calculated the transition probabilities and lifetimes, using the Cowan group of programs[27] run in the relativistic Hartree–Fock mode. An approximate function for the Hartree exchange potential was used, and the relativistic

mass correction and Darwin term were included in the self-consistent-field procedure. Scaled energy parameters were used in the calculations, the scaling factors being 0.85 and 0.95 for the electrostatic and spin-orbit integrals, respectively. The code also constructed and diagonalized energy matrices to provide energy eigenvalues and eigenvectors. To improve the agreement between experimental and calculated energies, we performed a least-squares fit of the energy parameters to the observed level values.

The even $5d^{10}$, $5d^9 6s$, and $5d^8 6s^2$ and the odd $5d^9 6p$ and $5d^8 6s 6p$ configurations were included in the least-squares fit. Unfortunately, only one of the experimental energy levels of Au II in Ref. 18 is assigned to the $5d^8 6s 6p$ configuration, which consists of 90 levels. That level, which appears among the $5d^9 7p$ levels, may be perturbed and is somewhat uncertain. We used the scaled Slater parameters for the $5d^8 6s 6p$ configuration to adjust the average energy to obtain agreement between the observed and calculated values of the only known level. The resulting Slater parameters were then used to calculate the transition probabilities for the various decays from the $5d^9 6p$ levels and the lifetimes of these levels. The theoretical data are given in Table 1.

To get an idea about the interaction between the $5d^9 6p$ and $5d^8 6s 6p$ configurations, we performed calculations for three different relative positions of these configurations. Besides the one discussed above, which is adjusted to the experimental energy levels given in Ref. 12, we successively lowered the energy of the whole $5d^8 6s 6p$ configuration by 5000 and 10 000 cm^{-1}. This resulted in an increase of the radiative lifetime of the $5d^9 6p$ levels by ~0.1 ns for each 5000-cm^{-1} step.

5. DISCUSSION

A study of Table 1 shows that the theoretical lifetimes of the $5d^9 6p$ levels are typically 30% shorter than the experimental values. There are a number of possible explanations for this discrepancy. The theoretical results contain uncertainties because the interaction between the levels of the $5d^9 6p$ and $5d^8 6s 6p$ configurations could be included in only a relatively crude way, since the $5d^8 6s 6p$ configuration is poorly known, as noted above. When one judges the quality of the experimental data, it is evident that line blends and cascades can give rise to uncertainties, unless properly analyzed. On the other hand, our measurement of the lifetime of the $6p\ ^2P_{1/2}$ level in Au I resulted in good agreement with accurate laser data, and this fact will increase the confidence in the experimental data for Au II.

To further investigate this problem, we also performed theoretical calculations of the f values of the $6s\ ^2S_{1/2}$–$6p\ ^2P_{1/2,3/2}$ resonance multiplet in Au I, using the same formalism as in the case of the Au II transitions, discussed above. The results obtained were $f = 0.241$ (1/2–1/2) and 0.474 (1/2–3/2). In an early paper Desclaux and Kim[28] studied this multiplet by means of several theoretical methods, obtaining $f = 0.631$ and 1.262 (nonrelativistic Hartree–Fock) and 0.296 and 0.623 (multiconfiguration Dirac–Fock). The latter results were in reasonable agreement with the calculations of Migdałek[29] (0.338 and 0.714). Some years later, Migdałek and Baylis[30,31] showed that the inclusion of core polarization in the dipole matrix element strongly affects the f values, resulting in the values 0.148 and 0.339 for the two lines.[31] Similar results were presented by Hafner and Schwarz,[32] who, using relativistic calculations with core polarization included, obtained the values 0.150 and 0.353. These data should be compared with the best experimental results, 0.176 ± 0.003 and 0.351 ± 0.015, reported by Hannaford *et al.*[26] Thus it is evident that the polarization of the $5d^{10}$ core can change the f values by a factor of ~2. In view of the fact that the present calculations for Au I deviate from the experimental values by ~30%, it would not be surprising if a similar effect occurred in the case of Au II.

Our conclusion is that the theoretical methods usually employed to calculate the f values for complex spectra such as Au II and Hg III are reliable to only ~30% and may need substantial refinements. More-accurate theoretical values can be obtained if one includes the interaction with the $5d$ electrons, but this requires a computational effort that would fall outside the present study. However, such studies are being initiated.

ACKNOWLEDGMENTS

We are grateful to T. Brage, C. Froese Fischer, and J. E. Hansen for valuable discussions and advice and J. F. Fuhr for informing us of recent studies of the lifetimes of gold ions. Two of the authors, R. Hellborg and I. Martinson, acknowledge the hospitality extended by the University of Toledo. R. Hellborg obtained travel support from the Royal Physiographic Society of Lund. L. J. Curtis and S. T. Maniak are supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences, under grant DE-FG05-88ER13958. The Swedish participants are supported by the Swedish Natural Sciences Research Council and the Swedish National Space Board.

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