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LIFETIME MEASUREMENTS IN Sn II

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ABSTRACT

Lifetime measurements are reported for levels arising from the $5s^25d$ and $5s^24f$ configurations in Sn II. Measured decay curves were jointly analyzed using the Arbitrarily Normalized Decay Curve (ANDC) method to remove the effects of cascade repopulation from the determination of the lifetimes of the $5s^25d$ $^2D_{3/2}$ level. The branching ratio of the decay of this level to the ground term fine-structure levels $^2P_{1/2}$ and $^2P_{3/2}$ was carefully measured, and we have obtained an accurate value for the absorption oscillator strength of the resonance transition to this level at 1400.52 Å. The results are discussed in the context of interpreting vacuum ultraviolet absorption spectra observed with the Goddard High Resolution Spectrograph on board the *Hubble Space Telescope*.

Subject headings: atomic data — ISM: abundances — ISM: atoms — methods: laboratory — ultraviolet: ISM

1. INTRODUCTION

Ultraviolet spectra acquired with the *Hubble Space Telescope* (*HST*) provided the first measures of elements beyond Zn in interstellar space (e.g., Cardelli et al. 1993; Cardelli 1994; Welty et al. 1995). The importance of elements beyond the iron group lies in their synthesis: production is governed by neutron capture and β-decay, while for lighter elements, processes involving α-particles dominate. For instance, nucleosynthesis of tin mainly involves slow neutron capture, which is believed to take place in helium-burning shells of asymptotic giant branch stars (Cameron 1982; Käppeler, Beer, & Wisshak 1989). Interstellar tin in its dominant interstellar form, Sn^+, was first detected through absorption at 1400 Å by Hobbs et al. (1993). In the most thorough study of the interstellar tin abundance to date, Sofia, Meyer, & Cardelli (1999) analyzed *HST* data for 14 lines of sight, which sample a range of diffuse environments.

A most interesting result of the survey by Sofia et al. (1999) is that they found sight lines having a tin abundance that appears to be greater than solar, a situation not known to exist for any other element. In this determination, Sofia et al. adopted the experimental oscillator strength of Andersen & Lindgård (1977) for the analysis. However, there are several reasons that a remeasurement of this oscillator strength seemed desirable. First, a recent theoretical calculation by Cardelli et al. (1993) produced *f*-values for selected heavy elements. That work resulted in predictions that agreed with experiments—where available—to within 10% to 20%, except for this transition, where the disagreement was almost a factor of 2. Second, the measurement of Andersen & Lindgård (1977) was carried out in the early days of beam-foil spectroscopy, when the effects of cascade repopulation—while known—were often insufficiently appreciated or taken into account. Finally, spin-orbit coupling was assumed to obtain an *f*-value from the measured lifetime, and the validity of that assumption needs to be investigated. We therefore undertook a new measurement, paying particular attention to the possibility that systematic effects were present. Cascade decay curves were measured and introduced into the analysis via the Arbitrarily Normalized Decay Curve (ANDC) method (Curtis, Berry, & Bromander 1971), and the branching ratio between the decays of the $^2D_{3/2}$ level to the $J = 3/2$ and $\frac{1}{2}$ lower levels was experimentally determined. Our new accurate laboratory measurements for the *f*-value of Sn II λ1400 place the conclusions of Sofia et al. (1999) on a more secure footing.

2. EXPERIMENT

This measurement utilized the University of Toledo Heavy Ion Accelerator Beam Foil Facility. Detailed descriptions of the experimental apparatus are provided in reports of earlier studies in this series (Schectman, Povolny, & Curtis 1998; Henderson et al. 1999) and in instrumentation reviews (Haar et al. 1993; Haar & Curtis 1993).

Ions of Sn^{2+} were produced by introducing pure tin into the Danfysik Model 911A Universal Ion Source. Extracted ions were accelerated through 20 kV and were magnetically analyzed. After momentum and mass-to-charge selection, the ions were postaccelerated through either 200 or 130 kV to obtain prefoil energies of 440 or 300 keV, respectively. The ions then entered an electrostatic switchyard and were steered into the beam-foil experimental station, where they were collimated before traversing thin carbon foils ranging in thickness from 2.1 to 2.5 $\mu\mathrm{g\ cm}^{-2}$. At these energies and foil thicknesses, charge state equilibrium occurs. The energies of the accelerated Sn^{2+} beams were chosen as compromises between obtaining the maximum yield of Sn^+ and having acceptable foil lifetimes and beam divergence due to Coulomb scattering in the foil. The observed spectra were quite clean, with no apparent overlap of the desired Sn II lines (McCormick & Sawyer 1938) with lines from the more abundant Sn III ions or the somewhat less populous Sn IV ions (A. G. Shenstone 1958, private communication quoted by Reader et al. 1980).

The Sn II emission lines were analyzed with an Acton 1 m normal incidence vacuum ultraviolet monochromator. For measurement of the transitions λ1400.52, λ1475.15, and λ1489.22, a 1200 l mm^{-1} grating blazed at 1500 Å was used with a solar blind photomultiplier tube. For the cascade transitions, viewed through the branches λ2448.98 and λ3283.21, a 600 l mm^{-1} grating blazed at 3000 Å was employed, along with a bi-alkali photodetector.

The postfoil velocity, determined from the accelerator calibration and estimates of the energy lost by the ions traversing the foil, was found to be 0.827 mm ns^{-1} at 440 keV and 0.678 mm ns^{-1} at 300 keV. The precision to which these velocities can be determined is limited by uncertainties in the energy calibration of the accelerator, uncertainties in foil thickness, and an imprecise knowledge of the details of the energy loss mechanism, especially the possible correlation between energy loss by nuclear scattering and direction. In no case, however, does the uncertainty with which the velocity is known exceed 2%. To minimize foil breakage during a run, the current through the foil was limited to 100 particle nA.

A preliminary fitting of all of the decay curves was carried out using a nonlinear least-squares multiexponential fitting program that is based on Marquardt's algorithm. Fits to the 4*f* and 5*f* decay curves were used as input to the ANDC analysis and also provide a lifetime for the $4f\ ^2F_{5/2}$ level. Even the somewhat heavily cascaded $5d\ ^2D_{3/2}$ decay curve met the criterion for extracting a reliable mean life by multiexponential fitting because its mean life was observed to be much shorter than those of the principal cascade transitions, which were also much weaker. These decay curves represent the dominant source of cascade repopulation of the 5*d* levels, and a cascade-correlated decay curve analysis method (Curtis et al. 1971) was also utilized to decrease the uncertainties in the determination of the lifetime of the $5d\ ^2D_{3/2}$ level. The overall uncertainties in our determinations were established by considering statistical uncertainties in the individual fits, scatter among the independent measurements, uncertainties in the beam velocity, and estimates of possible errors introduced by cascade corrections.

3. RESULTS

3.1. *Lifetimes*

Because the $5s^25p\ ^2P_{1/2}$–$5s^25d\ ^2D_{3/2}$ transition at 1400.52 Å is the main probe of Sn II abundance in the interstellar medium, our primary focus has been to make a precision measurement of the lifetime of the $5s^25d\ ^2D_{3/2}$ level and to obtain from it the absorption *f*-value for the above transition. Other lifetimes were by-products of this measurement and resulted either from the parametrization of the decay curves of cascade transitions for use in the ANDC analysis or from an attempt at assessing the extent to which *LS* coupling may be violated in this system. These lifetimes were not investigated with the same care and are not nearly as reliable. The lifetimes measured here are displayed in Table 1 along with a comparison with theory and with other measurements. Boldface in this table and in Table 2 highlight the new results presented here.

There are many factors that determine the ultimate precision of a beam-foil lifetime measurement. These include the accuracy to which the accelerator energy can be determined, the accuracy with which the foil thickness is known, the accuracy to which the energy lost in traversing the foil can be determined, the effects of multiple Coulomb scattering in causing the initially collimated beam to diverge, foil thickening, and effects of cascade repopulation. In order to check the importance of these phenomena, we have measured time-resolved decay curves for the $5s^25p\ ^2P_{1/2}$–$5s^25d\ ^2D_{3/2}$ transition for both 440 and 300 keV incident Sn^{2+} ions with respective beam velocities of 0.827 and 0.678 mm ns^{-1}. Most of the above effects depend strongly upon beam velocity, and comparing the two measurements allows us to estimate their importance.

The entrance slit and grating mask were configured to resolve, on the average, a 0.7 mm segment of the beam, which, for 440 keV Sn ions, corresponds to a nominal 0.8 ns time window viewed by the detection system. For this experimental setup, lifetimes significantly shorter than 0.8 ns approach the limit of determination by multiexponential curve-fitting alone. However, the ANDC method determines the primary lifetime from differences inherent in the exponential admixtures in the cascade and primary decay curves. It does not require that the primary exponential term be resolved, or even appear in the measured decay curve. Moreover, the good agreement between the values obtained from curve-fitting and from use of the ANDC method indicates that the lower limit has not yet been reached in this experiment. The excellent agreement between lifetimes extracted from multiexponential fits to the decay curves measured at two different velocities confirms this conclusion.

As described above, we measured decay curves for the principal cascade transitions that repopulate the $5d\ ^2D_{3/2}$ level. The cascade $5s^25d\ ^2D_{3/2}$–$5s^24f\ ^2F_{5/2}$ was studied in the alternate branch $5s5p^2\ ^2D_{3/2}$–$5s^24f\ ^2F_{5/2}$ at 3283.21 Å and the cascade $5s^25d\ ^2D_{3/2}$–$5s^25f\ ^2F_{5/2}$ was studied in the alternate branch $5s5p^2\ ^2D_{3/2}$–$5s^25f\ ^2F_{5/2}$ at 2448.98 Å. (The decay curve of the $5s^25f\ ^2F_{5/2}$ level was used in the ANDC analysis, but it was long-lived on this timescale and its lifetime is not reported here.) Another likely candidate for repopulating the $5s^25d\ ^2D_{3/2}$ level is the transition from the $5s^27p\ ^2P_{1/2}$ level. However, this transition is weak, and we

TABLE 1

LIFETIMES (IN NS)

Level	This Work	Other Experiments	Theory
$5s^25d\ ^2D_{3/2}$	**0.44 ± 0.02**	0.5 ± 0.05[a]	0.85,[b] 0.62,[c] 0.48,[d] 0.37[e]
$5s^24f\ ^2F_{5/2}$	**4.6 ± 1.0**	5.2 ± 0.5,[f] 5.0 ± 0.5,[g] 3.2 ± 0.5,[h] 9.0 ± 0.7[i]	3.2,[j] 3.1[k]
$5s^25d\ ^2D_{5/2}$	**0.46 ± 0.04**	...	0.50,[d] 0.42[e]

[a] Andersen & Lindgård 1977.

[b] Cardelli et al. 1993.

[c] Migdałek 1976.

[d] Marcinek & Migdałek 1994, J. Migdałek 2000 (private communication), SC.

[e] Marcinek & Migdałek 1994, CI assuming *LS* coupling.

[f] Gorshkov & Verolainen 1985.

[g] Alonso-Medina & Colón 2000 (sum of *A*-values).

[h] Wujec & Weniger 1977 and Wujec & Musielok 1976 (sum of *A*-values).

[i] Miller, Roig, & Bengtson 1979 (sum of *A*-values).

[j] Alonso-Medina & Colón 2000, relativistic Hartree Fock.

[k] Alonso-Medina & Colón 2000, intermediate coupling least-squares fit.

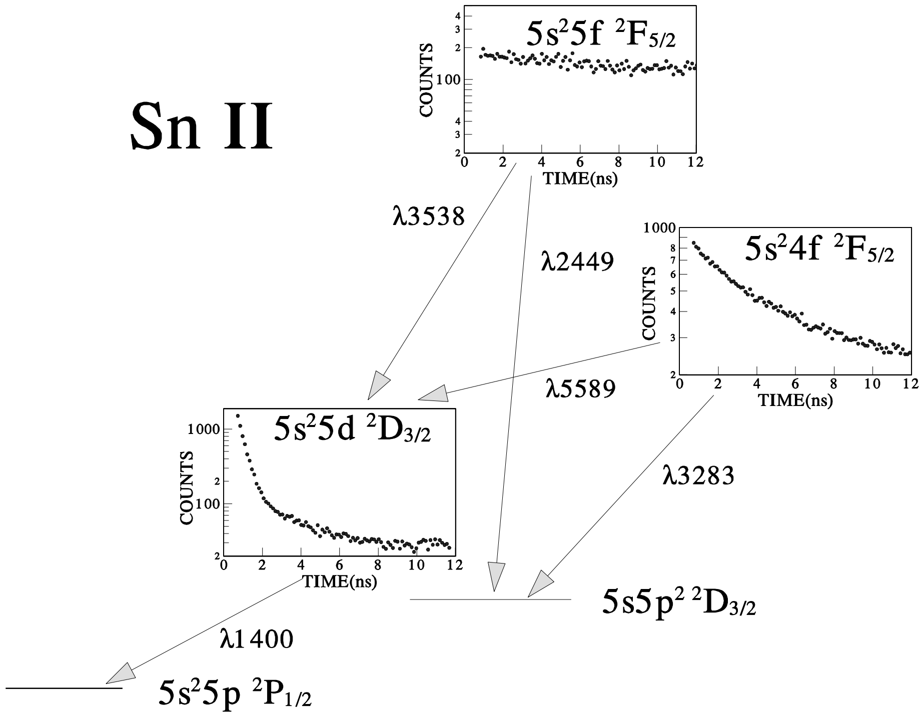

FIG. 1.—Schematic plot for Sn II of the decay curves of the primary $5s^2 5d\ ^2D_{3/2}$ level and its significant cascades, juxtaposed on a Grotrian diagram to illustrate their joint analysis by the ANDC method.

were unable to detect it. The measured transitions and their decay curves are shown in Figure 1. We carried out an ANDC cascade analysis to determine an accurate value for the mean life of the $5s^2 5d\ ^2D_{3/2}$ level. In addition, we determined this mean life by multiexponential curve-fitting, exploring a variety of ways of parametrizing the repopulation of the level at long times after excitation. Agreement between the various determinations of the lifetime was excellent and provided a further test of the precision of our result, $\tau = 0.44 \pm 0.02$ ns.

3.2. *The $5s^2 5p\ ^2P_{1/2}$ $5s^2 5d\ ^2D_{3/2}$ Absorption Oscillator Strength*

In order to extract the desired absorption *f*-value from the measured lifetime, it is also necessary to know the branching ratio between the transitions to the two fine-structure levels $J = 3/2$ and $J = \frac{1}{2}$ at 1489.22 Å and 1400.52 Å. We carefully measured this ratio to be 0.29 ± 0.02 (compared with the value expected if *LS* coupling were valid of 0.17). The measured ratio includes a correction for the relative efficiency of our detection apparatus, 0.76 ± 0.04, which was determined using an argon mini-arc secondary standard calibrated by the National Institute of Standards and Technology (NIST) (Bridges & Ott 1977). Combining the measured branching ratio with the measured lifetime gave an *A*-value for the transition at 1400.52 Å of $1.762 \pm 0.084 \times 10^9\ \mathrm{s}^{-1}$, with a corresponding *f*-value of 1.04 ± 0.05. It also follows that the *A*-value for the tran-

TABLE 2

BRANCHING FRACTIONS AND OSCILLATOR STRENGTHS (PARENTHESES DENOTE UNCERTAINTIES)

TRANSITION	λ[a] (Å)	BRANCHING FRACTION (%)	*f*		
			This Work	Other Measurements	Theory
$5s^2 5p\ ^2P_{1/2}$–$5s^2 5d\ ^2D_{3/2}$	1400.52	**78**[b]	**1.04(5)**	1.05(10)[c]	0.80,[d] 0.64,[e] 0.50,[f] 1.03,[g], 1.38[h]
$5s^2 5p\ ^2P_{3/2}$–	1489.22	**22**[b]	**0.170(14)**	...	0.086,[d] 0.108,[g] 0.130,[h]
$5s^2 5d\ ^2D_{3/2}$–$5s^2 4f\ ^2F_{5/2}$	5588.92	34[i]	0.51(1)[j]	...	...
$5s^2 5d\ ^2D_{5/2}$ –	5796.20	4[i]	0.044(1)[j]	...	...
$5s5p^2\ ^2D_{3/2}$–	3283.21	32[i]	0.169(4)[j]	...	...
$5s5p^2\ ^2D_{5/2}$–	3351.97	29[i]	0.106(2)[j]	...	...
$5s^2 5p\ ^2P_{3/2}$–$5s^2 5d\ ^2D_{5/2}$	1475.0	100	**1.06(9)**	...	0.74,[d] 0.97,[g] 1.18,[h]

[a] Air wavelengths are given for $\lambda > 2000$ Å.
[b] This work.
[c] Andersen & Lindgård 1977.
[d] Migdałek 1976.
[e] Cardelli et al. 1993.
[f] Andersen & Lindgård 1977, Coulomb approximation.
[g] Marcinek & Migdałek 1994, Migdałek 2000 (private communication), SC.
[h] Marcinek & Migdałek 1994, CI assuming *LS* coupling.
[i] Alonso-Medina & Colón 2000.
[j] Combines lifetime measured here with branching fraction from Alonso-Medina & Colón 2000.

sition at 1489.22 Å is $0.511 \pm 0.042 \times 10^9$ s^{-1}, leading to an *f*-value of 0.170 ± 0.014. The *f*-value determined for the $5s^25p\ ^2P_{1/2}$–$5s^25d\ ^2D_{3/2}$ transition confirms the value measured much earlier by Andersen & Lindgård (1977).

The results of our branching fraction and oscillator strength measurements are presented in Table 2 and are compared with other available measurements and theoretical calculations.

4. CONCLUDING REMARKS

The central results of this report are the accurate redetermination of the lifetime of the $5s^25d\ ^2D_{3/2}$ level in Sn II and the absorption *f*-value for the transition $5s^25p\ ^2P_{1/2}$–$5s^25d\ ^2D_{3/2}$ derived from it, displayed in Tables 1 and 2. The measured lifetime is in excellent agreement with the previous determination by Andersen & Lindgård (1977) and is of improved precision. The *f*-value obtained by combining our measured lifetime with our measured branching ratio is also in excellent agreement with that of Andersen & Lindgård, indicating that their determination was relatively insensitive to the assumption of *LS* coupling used in deriving it.

The state of the agreement with previous theoretical calculations is much less clear. The lifetimes predicted by the Coulomb approximation calculations of both Cardelli et al. (1993) and Andersen & Lindgård (1977) are nearly twice the experimental value. Migdałek's (Migdałek 1976) semiempirical calculation involving relativistic wave functions resulted in a predicted lifetime still some 40% too long. Later Marcinek & Migdałek (1994) presented theoretical multiplet oscillator strengths based upon Hartree-Fock-Pauli wave functions computed using the Cowan code (Cowan 1981). They carried out an extensive configuration interaction (CI) calculation as well as a simple single configuration (SC) calculation. All of these results are compared with our measurements in Tables 1 and 2. Marcinek & Migdałek's simple SC calculation agrees best with experiment, but their much more sophisticated CI calculation produces a lifetime nearly 30% too short. They initially suggested that perhaps cascade repopulation had affected the previous measurement by Andersen & Lindgård. However, our present study eliminates that possibility and suggests that further theoretical investigation is required. Until this situation is clarified, the case studied here remains an example of an atomic system for which reliance on experiment to determine reliable *f*-values is extremely important.

The conclusion of Sofia et al. (1999) that they had measured supersolar Sn II abundance along several sight lines of low molecular hydrogen density was based upon use of the experimental *f*-value of Andersen & Lindgård (1977). Since the use of the larger value obtained by Marcinek & Migdałek (1994) in their CI calculation would weaken this conclusion, it was important to reexamine the experimental situation. Our new result confirms the value of the oscillator strength assumed by Sofia et al. and makes it more precise. Using this value, they conclude that the interstellar tin abundance in low-density sight lines appears to be greater than the solar value, thus suggesting s-process enhancement.

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