# EXPERIMENTAL AND THEORETICAL RADIATIVE LIFETIMES, BRANCHING FRACTIONS, AND OSCILLATOR STRENGTHS FOR Lu I AND EXPERIMENTAL LIFETIMES FOR Lu II AND Lu III

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## ABSTRACT

Radiative lifetimes, accurate in most cases to  $\pm 5\%$ , from time-resolved, laser-induced fluorescence measurements on a slow beam of lutetium atoms and ions are reported for 22 odd-parity levels and 4 even-parity levels of Lu I and 14 odd-parity levels of Lu II. In addition, we report the radiative lifetime of one odd-parity level and an upper bound on the radiative lifetime of a second odd-parity level of Lu II. Experimental branching fractions for Lu I from emission spectra covering the near ultraviolet to the near infrared and recorded using the US National Solar Observatory 1.0 m Fourier transform spectrometer are reported. The branching fractions are combined with the radiative lifetimes to produce 44 experimentally determined transition probabilities or oscillator strengths, accurate generally to  $\pm 10\%$ , for Lu I. New theoretical values for Lu I radiative lifetimes and branching fractions from a relativistic Hartree-Fock calculation that includes core polarization effects are also reported. These experimental and theoretical results, as well as older published results, are compared.

Subject headings: atomic data — methods: laboratory — nuclear reactions, nucleosynthesis, abundances

#### 1. INTRODUCTION

There is an increasing need for accurate spectroscopic data, in particular oscillator strengths (f-values), on rare earth elements. Such data are essential in studies of chemically peculiar stars (Cowley & Mathys 1998), of "r" versus "s" nucleosynthesis of heavy elements (Sneden et al. 1995; Cowan et al. 1996; Sneden et al. 1996; Woolf, Tomkin, & Lambert 1995; Smith, Cunha, & Lambert 1995), and of other problems in stellar structure and evolution. Lines of Lu II, for example, have been identified in the extreme peculiar star HD 101065 (Cowley & Mathys 1998). Outside of astrophysics, the lighting-research community is interested in rare earth elements because of their rich emission spectra in the visible. Rare earth salts are used in many commercial metal-halide high-intensity discharge (HID) lamps. Accurate f-values are required in the models used for lamp design and for lamp diagnostics.

A combination of techniques from laser and Fourier transform spectroscopy has made it possible to measure large sets of accurate *f*-values (Lawler 1988). However, the synthesis of stellar spectra or of metal-halide HID lamp spectra must still depend on theoretical *f*-values to a substantial extent. Part of the motivation for this work is to explore further the utility of pseudo-relativistic Hartree-Fock calculations of spectroscopic data for rare earth elements.

In comparison with most lanthanides, lutetium has a rather simple spectrum. This is because it has only a few electrons, depending on the ionization stage, outside of the closed shell formed by the 14 f electrons. Lutetium is, therefore, a good test case for relativistic Hartree-Fock (HFR) calculations without overwhelmingly massive configuration interactions. The relativistic Hartree-Fock code used in this

work on Lu I was used in earlier work on Lu II (Quinet et al. 1999a) and Lu III (Biémont et al. 1999). It provided rather accurate results for Lu II. Here we extend the work to Lu I, which provides the challenge of three-valence electrons in a neutral atom. For comparison to the theoretical results, we present new measurements of radiative lifetimes, branching fractions, and oscillator strengths for Lu I. The radiative lifetimes are measured using the laser-induced fluorescence (LIF) apparatus at the University of Wisconsin (UW), and the branching fractions are determined using the 1.0 m Fourier transform spectrometer (FTS) at the US National Solar Observatory (NSO). Both of these experiments are discussed below. Once the LIF experiment was configured for work on Lu I, it involved only a slight additional effort to measure lifetimes for many levels in Lu II and even for a few levels in Lu III. These new experimental and theoretical results, as well as older published results, are compared.

## 2. LIFETIME MEASUREMENTS

Radiative lifetimes for 22 odd-parity and 4 even-parity levels of Lu I, 14 odd-parity levels of Lu II, and 2 odd-parity levels of Lu III were measured using time-resolved LIF on a slow atom/ion beam produced from a pulsed hollow cathode discharge (HCD). The apparatus is the same as that recently used to determine radiative lifetimes for Ho I and Ho II (Den Hartog, Wiese, & Lawler 1999). Details of the experiment, including an in-depth discussion of all the known systematic errors, are given in Den Hartog et al. (1999).

A weakly collimated slow beam of lutetium atoms and ions is generated in a large-bore HCD lined with lutetium metal. The discharge is maintained with 0.3–0.4 torr of argon buffer gas and about 60 mA of DC current. A current

pulse with a peak of 5–15 A and a duration of 10  $\mu$ s is applied to the HCD and is timed to occur just before the dye laser pulse. Many metastable states are populated in the atom/ion beam. The atom/ion beam enters a vacuum scattering chamber ( $\sim 10^{-4}$  torr background pressure) where it intersects a pulsed laser beam at a right angle. The laser beam enters and exits the scattering chamber through a pair of fused silica Brewster windows. The laser system consists of a two-stage dye laser, constructed at UW, pumped by a pulsed N<sub>2</sub> laser operated at 30 Hz. The oscillator cavity is defined on one side of the first dye cell by a wedged uncoated window, which serves as the output mirror, and on the other side by a prismatic beam expander followed by an echelle diffraction grating mounted in a pressure chamber. The angle of the grating and chamber pressure provide course and fine-tuning of the laser, respectively. The laser output of the oscillator cavity passes through a second dye cell, which serves as the amplifier. The dye laser has a pulse duration of about 3 ns (FWHM) and a bandwidth of about  $0.2 \text{ cm}^{-1}$  (FWHM). A selection of dyes and frequency doubling crystals enables the dye laser to be tuned over a wavelength range of 205-720 nm. A 0.5 m focal length monochromator is used to measure the laser wavelength to within  $\pm 0.05$  nm after the laser beam exits the scattering chamber.

The laser is tuned to a transition from the ground or metastable state to the upper level of interest. The laserinduced fluorescence from the excited level is focused onto a photomultiplier tube (PMT) by an f/1 optical system orthogonal to the laser and ion/atom beams. The optical system consists minimally of two fused silica lenses and an RCA 1P28A PMT. Spectral filters are often placed between the lenses to prevent fluorescence from cascades through lower levels or from line blends from reaching the PMT. Since levels are selectively excited by the laser, cascades from higher lying levels are not an issue. For long lifetimes ( $\tau > 100$  ns for ions and  $\tau > 300$  ns for neutral atoms), an additional plano-convex cylindrical lens is used to defocus the fluorescence light at the PMT, making the optical system less sensitive to the atomic motion that would otherwise tend to artificially shorten the measured lifetime. For very long lifetimes,  $\tau > 1 \mu s$ , the additional lens is not sufficient to completely avoid this systematic effect. A correction factor is obtained using the time-of-flight selection of slow atoms, as described by Marsden et al. (1988) and also discussed in Den Hartog et al. (1999) and Curry, Den Hartog, & Lawler (1997). In this work, only two radiative lifetimes of Lu I were greater than 1  $\mu$ s.

A Tektronix SCD1000 transient digitizer is used to record the fluorescence signal from the PMT. Each digitized trace represents an average of 640 decay curves. Background signals are determined by tuning the laser slightly off of the fluorescence transition. These are digitally subtracted from the fluorescence signal. Scattered light from the laser is reduced in the fluorescence signal by discarding at least the first 7 ns after the peak of the laser pulse. The time-resolved fluorescent decay curve is separated into two analysis regions along the time axis. Each analysis region represents a time window roughly 1.5 times the lifetime in length. A single exponential is fitted to the backgroundsubtracted signal in each analysis region to determine the lifetime. Consistency between the lifetimes determined from the two analysis regions provides a check on systematic effects such as cascades, blends, or electronic artifacts that may distort the decay curve. A single measurement is an average of the lifetimes determined from five backgroundsubtracted traces. Statistical errors are less than 3% for the weakest lines and are generally much less than 1%. A minimum of two measurements, taken at least several days apart, are recorded for each lifetime reported. Whenever possible, lifetime measurements are made by using two different transitions to excite the atom or ion to the upper level of interest. The agreement of lifetime measurements using two different lines for laser excitation of the upper level provides an extra degree of confidence in the original line classifications, in the line identifications of this experiment, and in the absence of line blending.

Zeeman quantum beats caused by the precession of atoms in a magnetic field can distort the fluorescence signal and thus be a possible systematic error. For lifetime measurements less than 300 ns, the magnetic field within the scattering chamber is zeroed to within 0.02 G. This tolerance is not sufficient for measurement of longer lifetimes. Therefore, for  $\tau > 300$  ns, a high magnetic field is applied to the scattering chamber (~30 G) to cause rapid oscillations that are effectively averaged out during the timescales of interest.

Owing to the relatively simple spectra of Lu I and Lu II, line identifications are not difficult. The first step is to tune the dye laser to within 0.1 nm of the desired transition by changing the angle of the laser grating. For line identifications, we record the fluorescence spectra over a 0.5-1.0 nm range by scanning the pressure in the dye laser. The position and wavelength of lines appearing in the spectra can be determined very precisely and compared to the Lu line-list of Meggers, Corliss, & Scribner (1975). Line identifications are extremely reliable when two or more identifiable lines appear in the fluorescence spectra.

The range of lifetimes that can be measured in the apparatus is limited on the long side by the length of time the atom or ion spends in the scattering chamber. All the lifetimes reported here are much less than this limit, which is about 4  $\mu$ s. The fidelity and overall electronic bandwidth of the detection system limit the shortest measurable lifetime to about 1.8 ns. Many of the Lu lifetimes reported here are short; however, we periodically check our systematic errors by measuring accurate, well-known radiative lifetimes. These are discussed in Den Hartog et al. (1999) and are also summarized in Nitz, Bergeson, & Lawler (1995) and in Curry et al. (1997). Accurate calculations for Be I (Weiss 1995) and measurements for Fe II (Guo et al. 1992; Biémont et al. 1991) and Cu I (Carlsson, Sturesson, & Svanberg 1989) allow cross-checks for lifetimes in the range of 1.8-8 ns. Thus we have a great deal of confidence in our short lifetimes, which are accurate to the greater of  $\pm 5\%$  or  $\pm 0.2$  ns. Precise He I calculations (Kono & Hattori 1984) give reliable cross-checks for radiative lifetimes in the range of 95–220 ns. Our longer lifetimes ( $\tau > 4$  ns) are generally good to within 5%. For the Lu experiment, cross-checks were performed before Lu measurements were begun, when roughly half the measurements were complete, and after the measurements were complete.

Our radiative lifetime measurements for Lu I are summarized in Table 1. New relativistic Hartree-Fock calculations, discussed in § 4, are also presented in Table 1. Our radiative lifetime measurements for Lu II and Lu III are presented in Tables 2 and 3, respectively. Previously reported experimental and theoretical values for Lu I, Lu II, and

				Experimentai	LIFETIME (ns)	THEORETICAL LIFETIMES (ns)				
Configuration	Term	J	Upper Level (cm <sup>-1</sup> )	This Experiment	Other Experiments	HFR (A)	HFR (B)	HFR (C)	HFR (D)	HFR (E)
5d6s ( <sup>3</sup> D) 6p	${}^{4}F^{o}$	1.5	17427.28	554 ± 28		370	451	398	599	649
$5d6s (^{3}D) 6p \dots$	${}^{4}F^{o}$	2.5	18504.58	$472 \pm 24$	$430 \pm 20^{a}$	303	364	331	499	520
$5d6s (^{3}D) 6p \dots$	$^{4}D^{o}$	0.5	20762.47	$1050 \pm 52$	$1020 \pm 60^{a}$	693	833	761	1180	679
$5d6s (^{3}D) 6p \dots$	$^{4}D^{o}$	1.5	21195.37		$2450 \pm 150^{a}$					
$5d6s (^{3}D) 6p \dots$	$^{2}D^{o}$	2.5	21462.38	81.9 ± 4.1	$80 \pm 4^{a}$	52.5	64.1	55.8	83.9	98.5
5d6s ( <sup>3</sup> D) 6p	$\frac{1}{2}D^{o}$	 1.5	 22124.76	 43.9 ± 2.2	$83 \pm 5^{b}$ $43 \pm 3^{a}$	 25.2	 28.7	 27.1	 40.6	 
				•••	$47 \pm 1^{b}$					
				•••	$48.8 \pm 1.0^{\circ}$	•••		•••	•••	•••
$5d6s (^{3}D) 6p \dots$	$^{4}D^{o}$	2.5	22221.68	862 ± 43	$820 \pm 50^{a}$	305	285	332	497	
$5d6s (^{3}D) 6p \dots$	${}^{4}P^{o}$	1.5	24308.09	$1640~\pm~131$		667	725	695	1050	
5d6s ( <sup>3</sup> D) 6p	${}^{4}P^{o}$	2.5	25191.56	$332 \pm 17$	$315 \pm 25^{b}$	202	229	219	327	
$5d6s (^{1}D) 6p \dots$	${}^{2}F^{o}$	2.5	28020.11	$10.5 \pm 0.5$	$12 \pm 1^{b}$	9.3	9.2	10.8	15.7	23.2
			•••		$12 \pm 1^{d}$					
$6s^2$ ( <sup>1</sup> S) $7p$	${}^{2}P^{o}$	0.5	29431.05	$9.2 \pm 0.5$	$8.6 \pm 0.8^{b}$	9.6	9.6	10.9	13.1	13.4
5d6s ( <sup>1</sup> D) 6p	${}^{2}F^{o}$	3.5	29486.94	$151 \pm 8$	$165 \pm 5^{b}$	114	76.7	133	187	
$5d6s (^{1}D) 6p \dots$	${}^{2}D^{o}$	1.5	29608.01	$5.9 \pm 0.3$	$7.2 \pm 0.3^{b}$	8.0	7.9	9.0	12.6	9.4
					$5.12 \pm 0.16^{\circ}$					
$5d6s (^{3}D) 6p \dots$	${}^{2}F^{o}$	2.5	30183.55	$5.3 \pm 0.3$	$6.7 \pm 0.4^{b}$	4.5	4.8	5.2	7.5	
$6s^2$ ( <sup>1</sup> S) $7p$	${}^{2}P^{o}$	1.5	30488.62	$5.9 \pm 0.3$	$7.4 \pm 0.6^{b}$	5.8	7.2	6.6	8.9	8.7
$5d6s (^{1}D) 6p \dots$	${}^{2}P^{o}$	1.5	31523.03	$8.5 \pm 0.4$	$10.0 \pm 0.5^{b}$	6.7	8.2	7.8	10.7	9.0
$5d6s (^{3}D) 6p \dots$	${}^{2}F^{o}$	3.5	31751.17	$4.1 \pm 0.2$	$5.7 \pm 0.4^{b}$	3.3	3.4	3.8	5.5	5.0
$5d6s(^{3}D)6p$	${}^{2}P^{o}$	0.5	32058.10	$4.9 \pm 0.2$	$6.3 \pm 0.4^{b}$	5.8	5.9	6.7	9.5	7.8
$5d6s (^{1}D) 6p \dots$	$^{2}D^{o}$	2.5	32456.70	$3.2 \pm 0.2$	$5.1 \pm 0.6^{b}$	3.0	3.4	3.5	4.9	
$5d6s(^{1}D)6p$	${}^{2}P^{o}$	0.5	33443.10	$7.8 \pm 0.4$	$9.1 \pm 0.6^{b}$	4.1	5.8	4.6	6.9	7.3
$5d6s(^{3}D)6p$	${}^{2}P^{o}$	1.5	34436.49	3.7 + 0.2	$5.1 + 0.3^{b}$	3.2	3.4	3.6	5.2	5.5
$6s^2$ ( <sup>1</sup> S) $5f$	${}^{2}F^{o}$	2.5	36633.36	30.2 + 1.5	$31 + 2^{b}$	31.0	52.9	31.6	35.7	32.9
$6s^2$ ( <sup>1</sup> S) $5f$	${}^{2}F^{o}$	3.5	36644.12	30.3 + 1.5	$29 + 2^{b}$	33.0	56.1	33.8	38.7	36.7
$6s^2$ ( <sup>1</sup> S) 7s	$^{2}S$	0.5	24125.99	$12.3 \pm 0.6$	-	12.4	11.8	11.7	11.7	11.9
$5d^2$ ( <sup>3</sup> <i>P</i> ) 6 <i>s</i>	$^{2}P$	0.5	28793.42	45.7 + 2.3		68.6	102.3	56.5	71.9	
$6s^2$ ( <sup>1</sup> S) 6d	$^{2}D$	1.5	31542.24	9.2 + 0.5	$20 + 2^{b}$	10.4	8.8	10.7	10.5	10.7
$6s^2$ ( <sup>1</sup> S) $6d$	$^{2}D$	2.5	31713.60	$11.2 \pm 1.1$	$19 \pm 2^{\mathrm{b}}$	14.8	12.5	15.2	14.9	15.1

 TABLE 1

 Experimental and Theoretical Radiative Lifetimes for Lu i

NOTE.—Details of the various HFR calculations are discussed in the text, as well as the reason for omission of certain results from calculation HFR (E).

<sup>a</sup> Kwiatkowski et al. 1980.

<sup>b</sup> Gorshkov et al. 1984.

° Göbel 1970a.

<sup>d</sup> Göbel 1970b.

° Göbel 1971.

Lu III are also presented in Tables 1, 2, and 3 and are discussed in  $\S$  5.

### 3. BRANCHING FRACTION MEASUREMENTS

Branching fractions for Lu I in the visible and near-UV were measured using spectra taken with the 1.0 m FTS at the NSO. Since the Lu I data is obtained from the same data sets used to determine the previously published Lu II branching fractions (Quinet et al. 1999a), the reader is referred to that publication for details of the experiment. A few branching fractions were also measured using an Acton Research Corp. 1.0 m focal length grating spectrometer equipped with a 2400 line  $mm^{-1}$  holographic grating. In both experiments, two sealed, commercially available, lutetium-lined HCD lamps were used as a source of Lu I emission lines. One of the lamps contained neon as buffer gas while the other used argon. Five FTS spectra at four discharge currents were taken with the Ar-Lu lamp: two spectra at 30 mA, and one each at 15, 12, and 10 mA. Two spectra, one with 15 and the other with 25 mA of discharge current, were recorded using the Ne-Lu lamp. The Ne-Lu spectra is used to check the Ar-Lu spectra for blends.

The HCD lamps used in this study operate with relatively low buffer gas pressures and are not in local thermodynamic equilibrium (LTE). This is not a problem because branching fractions are determined for transitions with common upper levels. Because of the low collision rate, Doppler broadening tends to dominate the emission-line shape. Radiation trapping effects can be identified and corrected by comparing the spectra taken at different discharge currents.

When measuring emission branching fractions, determining the radiometric calibration or efficiency is critical to the experiment. Detectors, spectrometer optics, lamp windows, and any other components in the light path or any reflections which contribute to the detected signal (such as that caused by light reflecting off the back of the HCD) all have wavelength-dependent optical properties that must be taken into account when determining the ratio of line intensities at different wavelengths. An excellent way to deter-

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EXPERIMENTAL AND THEORETICAL RADIATIVE LIFETIMES FOR LU II

				LIFETIME (ns)		
Configuration	Term	J	$\frac{\text{UPPER LEVEL}}{(\text{cm}^{-1})}$	This Experiment	Other Experiments	Theory
6s6p	${}^{3}P^{o}$	0	27264.40	64.8 ± 3.2	$61 \pm 5^a$	69.66 <sup>ь</sup>
6s6p	${}^{3}P^{o}$	1	28503.16	$37.4 \pm 1.9^{\circ}$	$37 \pm 4^{a}$	37.60 <sup>b</sup>
6s6p	${}^{3}P^{o}$	2	32453.26	37.6 ± 1.9	$42 \pm 6^{a}$	46.47 <sup>b</sup>
6s6p	${}^{1}P^{o}$	1	38223.49	$2.3 \pm 0.2$		
5d6p	${}^{3}F^{o}$	2	41224.96	$5.9 \pm 0.3$	$3.7 \pm 0.4^{d}$	5.31 <sup>b</sup>
-						5.18°
5d6p	${}^{3}F^{o}$	3	44918.68	$4.8 \pm 0.2$	$5.09 \pm 0.18^{b}$	4.45 <sup>⊾</sup>
-					$3.8 \pm 0.5^{\rm f}$	4.21°
5d6p	${}^{1}D^{o}$	2	45458.56	$4.3 \pm 0.2$	$2.8 \pm 0.3^{d}$	3.86 <sup>b</sup>
•						3.92°
5d6p	$^{3}D^{o}$	1	45532.33	$3.3 \pm 0.2$	$3.1 \pm 0.2^{a}$	2.81 <sup>b</sup>
•					$2.4 \pm 0.3^{\rm f}$	2.51°
5d6p	$^{3}D^{o}$	2	46904.38	$3.5 \pm 0.2$	$3.8 \pm 0.5^{\rm f}$	3.05 <sup>b</sup>
•						3.01°
5d6p	${}^{3}F^{o}$	4	48536.83	$3.9 \pm 0.2$	$3.96 \pm 0.16^{b}$	3.45 <sup>⊾</sup>
•				-	$4.2 + 0.4^{\rm f}$	3.25°
5d6p	$^{3}D^{o}$	3	48733.19	$3.4 \pm 0.2$	$3.80 \pm 0.18^{b}$	3.06 <sup>b</sup>
•				-	$4.0 + 0.5^{\rm f}$	2.91°
5d6p	<sup>3</sup> P <sup>o</sup>	0	49963.58	2.5 + 0.2	-	2.05 <sup>g</sup>
5d6p	<sup>3</sup> P <sup>o</sup>	1	50049.20	2.5 + 0.2	$3.4 + 0.5^{f}$	2.07 <sup>b</sup>
1				-	-	1.94°
5d6p	${}^{3}P^{o}$	2	51201.66	$2.6 \pm 0.2$		2.16 <sup>g</sup>
5d6p	${}^1F^o$	3	53079.33	$4.4 \pm 0.2$	$4.50 \pm 0.22^{b}$	3.96 <sup>b</sup>

<sup>a</sup> Li & Lundberg 1998.

<sup>b</sup> Quinet et al. 1999a.

° Den Hartog et al. 1998.

<sup>d</sup> Andersen et al. 1975.

<sup>e</sup> Bord et al. 1998.

<sup>f</sup> Andersen & Sorensen 1974.

<sup>g</sup> Unpublished result from HFR calculation described by Quinet et al. 1999a.

mine the relative radiometric efficiency of an FTS is to compare well-known branching ratios for sets of lines well separated in wavelength to the intensities measured for the same lines. Sets of Ar I and Ar II lines have been established for this purpose in the range of 4300–35000 cm<sup>-1</sup> by Whaling, Carle, & Pitt (1993), Hashiguchi & Hasikuni (1985), Danzmann & Kock (1982), and by Adams & Whaling (1981). These provide an excellent means of calibrating the spectra since the argon lines are measured in the exact experimental arrangement and at the exact same time as were the Lu I lines.

Branching fractions from the 5d6s (<sup>1</sup>D) 6p <sup>2</sup>D<sup>o</sup><sub>5/2</sub> level at 32456.70 cm<sup>-1</sup> and from the 5d6s (<sup>3</sup>D) 6p <sup>2</sup>P<sup>o</sup><sub>3/2</sub> level at 34436.49 cm<sup>-1</sup> of Lu I were measured using the 1 m focal length grating spectrometer. These levels have significant branches in the UV beyond the limit of the FTS spectra. A

argon mini-arc (Bridges & Ott 1977), calibrated at the US National Institute of Standards and Technology (NIST), was used to radiometrically calibrate the grating spectrometer. All the experimentally determined branching fractions are presented in Table 4, along with those determined from the HFR calculations, and discussed in the following section.

### 4. HFR CALCULATIONS

For heavy neutral atoms such as Lu I, both intravalence and core-valence correlation have to be taken into account in atomic structure calculations. Simultaneous treatment of these effects within a configuration interaction (CI) scheme is very complex and reliable only if enough configuration mixing is considered. However, in practice, even a large computer imposes rather severe limitations on the number

TABLE 3								
Experimental and Theoretical Radiative Lifetimes for Lu III								
LIFETIME (ns)								
Configuration	Term	J	Upper Level (cm <sup>-1</sup> )	This Experiment		Theory		
$\frac{4f^{14} ({}^{1}S) 6p \dots}{4f^{14} ({}^{1}S) 6p \dots}$	<sup>2</sup> P <sup>o</sup> <sup>2</sup> P <sup>o</sup>	0.5 1.5	38400.61 44705.21	$2.1 \pm 0.2 < 1.8$	2.23ª 1.47ª	2.19 <sup>ь</sup> 1.46 <sup>ь</sup>	2.54° 1.66°	

<sup>a</sup> Biémont et al. 1999.

<sup>b</sup> Migdalek 1982, using  $\alpha_d = 5.20a_0^3$ 

° Migdalek 1982, using  $\alpha_d = 8.20a_0^3$ .

TABLE 4								
EXPERIMENTAL AND	THEORETICAL BRANCHING	FRACTIONS FOR	Luı					

TRANSITION				BRANCHING FRACTION		
Wavenumber (cm <sup>-1</sup> )	Lambda in Air (nm)	Upper-Level Energy Parity $J$ (cm <sup>-1</sup> )	Lower-Level Energy Parity $J$ (cm <sup>-1</sup> )	This Experiment	HFR (D)	
17427.28	573.654	17427.28 od 1.5	0.00 ev 1.5	$0.960 \pm 0.010$	0.964	
15433.36	647.768	17427.28 od 1.5	1993.92 ev 2.5	$0.040 \pm 0.003$	0.036	
18504.56	540.257	18504.58 od 2.5	0.00 ev 1.5	$0.566 \pm 0.007$	0.556	
16510.64	605.502	18504.58 od 2.5	1993.92 ev 2.5	$0.434 \pm 0.007$	0.444	
18438.61	542.190	20432.53 od 3.5	1993.92 ev 2.5	$1.000 \pm 0.020$	0.999	
20762.42	481.505	20762.47 od 0.5	0.00 ev 1.5	$0.950 \pm 0.038$	0.954	
21195.37	471.669	21195.37 od 1.5	0.00 ev 1.5	$0.277 \pm 0.025$	0.360 <sup>a</sup>	
19201.45	520.649	21195.37 00 1.5 21462.28 od 2.5	1993.92 eV 2.5	$0.623 \pm 0.051$	0.537	
21402.33	405.802	21402.38 00 2.5 21462.38 od 2.5	0.00 eV 1.5 1003 02 eV 2.5	$0.238 \pm 0.006$ $0.742 \pm 0.006$	0.232	
22124 70	451 857	21402.38 Ou 2.3	0.00 ev 1.5	$0.742 \pm 0.000$	0.708	
20130 78	496 613	22124.76 od 1.5	1993 92 ev 2 5	$0.0014 \pm 0.0002$	0.0062ª	
22221.64	449.886	22221.68 od 2.5	0.00  ev  1.5	$0.110 \pm 0.0002$	0.106	
20227.72	494.233	22221.68 od 2.5	1993.92 ev 2.5	$0.840 \pm 0.034$	0.866	
24308.20	411.268	24308.09 od 1.5	0.00 ev 1.5	0.706 + 0.071	0.875ª	
22314.28	448.018	24308.09 od 1.5	1993.92 ev 2.5	$0.144 \pm 0.015$	0.081ª	
25191.57	396.846	25191.56 od 2.5	0.00 ev 1.5	$0.732 \pm 0.030$	0.668	
23197.65	430.957	25191.56 od 2.5	1993.92 ev 2.5	$0.218 \pm 0.010$	0.313ª	
28020.18	356.784	28020.11 od 2.5	0.00 ev 1.5	$0.721 \pm 0.011$	0.690	
26026.26	384.118	28020.11 od 2.5	1993.92 ev 2.5	$0.278 \pm 0.009$	0.309	
29430.90	339.681	29431.05 od 0.5	0.00 ev 1.5	$0.950 \pm 0.039$	0.932	
2/493.02	363.625	29486.94 od 3.5	1993.92 ev 2.5	$0.9/0 \pm 0.020$	0.975	
29607.98	337.030	29608.01 0d 1.5 20182.55 od 2.5	0.00 eV 1.5	$0.990 \pm 0.010$	0.961	
28180.63	351.211	30183.55 od 2.5	1003 02 ev 2.5	$0.981 \pm 0.010$ 0.0130 $\pm 0.0022$	0.994	
30488.62	327 897	30488 62 od 1 5	0.00 ev 1.5	$0.0139 \pm 0.0022$ 0.590 + 0.011	0.005	
28494 70	350 842	30488.62 od 1.5	1993 92 ev 2 5	$0.390 \pm 0.011$ 0.385 + 0.008	0.404	
31523.14	317.136	31523.03 od 1.5	0.00  ev 1.5	$0.562 \pm 0.000$	0.749	
29529.22	338.550	31523.03 od 1.5	1993.92 ev 2.5	0.428 + 0.009	0.238	
29757.25	335.956	31751.17 od 3.5	1993.92 ev 2.5	$0.999 \pm 0.007$	0.999	
32058.10	311.843	32058.10 od 0.5	0.00 ev 1.5	0.995 ± 0.021	0.988	
32456.87	308.012	32456.70 od 2.5	0.00 ev 1.5	$0.0229 \pm 0.0011$	0.021ª	
30462.95	328.173	32456.70 od 2.5	1993.92 ev 2.5	$0.976 \pm 0.010$	0.978	
33443.20	298.927	33443.10 od 0.5	0.00 ev 1.5	$0.995 \pm 0.005$	0.938	
34436.49	290.305	34436.49 od 1.5	0.00 ev 1.5	$0.0292 \pm 0.0011$	0.039	
32442.57	308.147	34436.49 od 1.5	1993.92 ev 2.5	$0.9/1 \pm 0.010$	0.937	
30033.31	2/2.895	30033.30 00 2.3 36644.12 ad 2.5	0.00 eV 1.5	$0.700 \pm 0.049$	0.510	
18331 53	200.314 545 357	22467 56 ev 1 5	4136 13 od 0 5	$0.700 \pm 0.049$ 0.806 + 0.030	0.370	
14991.18	666.875	22467.56 ev 1.5	7476.39 od 1.5	$0.000 \pm 0.030$ $0.194 \pm 0.030$	0.270	
19989.86	500.114	24125.99 ev 0.5	4136.13 od 0.5	$0.393 \pm 0.010$	0.465	
16649.51	600.452	24125.99 ev 0.5	7476.39 od 1.5	0.607 + 0.010	0.535	
20382.16	490.488	24518.30 ev 1.5	4136.13 od 0.5	$0.968 \pm 0.010$	0.963	
17041.81	586.630	24518.30 ev 1.5	7476.39 od 1.5	$0.0218 \pm 0.0016$	0.033ª	
24657.34	405.444	28793.42 ev 0.5	4136.13 od 0.5	$0.919 \pm 0.015$	0.904	
21316.99	468.978	28793.42 ev 0.5	7476.39 od 1.5	$0.061 \pm 0.006$	0.077 <sup>a</sup>	
25801.81	387.460	29937.90 ev 1.5	4136.13 od 0.5	$0.126 \pm 0.015$	0.184ª	
22401.40 26611.10	445.082	29937.90 ev 1.5	/4/0.39 od 1.5	$0.750 \pm 0.055$	0./16	
20011.18	3/3.0/3	30747.30 eV 0.5	4136.13 00 0.5 7476.20 od 1.5	$0.349 \pm 0.014$	0.319	
25270.65	429.002	30747.30 eV 0.3	/4/0.59 00 1.5	$0.021 \pm 0.018$ 0.845 $\pm 0.006$	0.007	
2/400.24	<i>415 4</i> 09	31542.24 EV 1.5 31542.24 EV 1.5	7476 39 od 1 5	$0.843 \pm 0.000$ 0.155 + 0.006	0.004	
24237.25	412 472	31713.60 ev 2.5	7476 39 od 1 5	1.000 + 0.000	0.990	
25510.40	391.886	32986.62 ev 0.5	7476.39 od 1.5	$0.118 \pm 0.014^{b}$	0.048	
12224.33	817.816	32986.62 ev 0.5	20762.47 od 0.5	$0.325 + 0.041^{b}$	0.341	
11791.38	847.844	32986.62 ev 0.5	21195.37 od 1.5	$0.297 + 0.038^{b}$	0.351	
12636.17	791.161	33831.46 ev 1.5	21195.37 od 1.5	$0.177 \pm 0.017^{b}$	0.152	
11609.90	861.097	33831.46 ev 1.5	22221.68 od 2.5	$0.478 \pm 0.022^{b}$	0.470	
9722.82	1028.226	33831.46 ev 1.5	24108.72 od 0.5	$0.129 \pm 0.017^{b}$	0.162	
27798.20	359.633	35274.50 ev 2.5	7476.39 od 1.5	$0.153 \pm 0.025^{b}$	0.082	
16769.99	596.138	35274.50 ev 2.5	18504.58 od 2.5	$0.035 \pm 0.006^{b}$	0.019	
11750.31	850.808	35274.50 ev 2.5	23524.24 od 3.5	$0.368 \pm 0.020^{\text{b}}$	0.372	
13812.20	/23./98	35274.50 ev 2.5	21462.38 od 2.5	$0.107 \pm 0.015^{\circ}$	0.126	
10900.33	911.030	55274.50 eV 2.5	243U8.U9 00 1.3 25101 56 ad 2.5	$0.110 \pm 0.011^{\circ}$	0.141	
10002.70	771.470	JJZ 14.JU EV 2.J	20191.00 OU 2.0	$0.179 \pm 0.010^{-1}$	0.219	

<sup>a</sup> Cancellation effects are important in these theoretical branching fractions. The CF or cancellation factor (Cowan 1981) is less than 0.05. <sup>b</sup> The experimental branching fractions from these levels have been corrected for IR lines beyond the limit of the spectra using theoretical results reported herein.

of interacting configurations that can be considered simultaneously and, in particular, the inclusion of corepolarization effects through consideration of a huge number of configurations with open shells is often prevented by computer limitations. This is particularly true in the case of Lu I, which is characterized by a  $Lu^{+3}$  core surrounded by three-valence electrons.

In our work, we have used an approach in which most of the intravalence correlation is represented within a configuration superposition scheme while core-valence correlation is represented approximately by considering a corepolarization model potential. Hibbert (1982a) has discussed a variety of different forms for this core-polarization potential. In this paper, we have adopted the form introduced by Migdalek & Baylis (1978) and used extensively in recent calculations for atoms and ions with one- or two-valence electrons (see, e.g., Biémont et al. 1998a, 1999; Biémont, Quinet, & Van Renterghem 1998b; Palmeri et al. 2000a, 2000b; Quinet et al. 1999a; Quinet, Palmeri, & Biémont 1999b).

Outer correlation has been retained among the configurations  $5d6s^2$ ,  $6s^2$  ns (n = 7-10),  $6s^2$  nd (n = 6-10),  $6s6p^2$  $5d^{2}6s$ ,  $5d^{2}7s$ ,  $5d^{2}6d$ ,  $5d^{2}7d$ , 5d6s7s, 5d6s6d, 5d6s7d,  $5d6p^{2}$ ,  $5d^3$  (even parity) and  $6s^2 np$  (n = 6-10),  $6s^2 nf$  (n = 5-10),  $5d^26p, 5d^27p, 5d^25f, 5d^26f, 5d^27f, 5d6s6p, 5d6s7p, 5d6s5f,$ 5d6s6f, 5d6s7f (odd parity). Using a well-established leastsquares fitting procedure (Cowan 1981), the radial parameter values were adjusted to obtain the best agreement between the calculated and the experimental energy levels below 40,000 cm<sup>-1</sup> taken from Vergès & Wyart (1978) and from the NIST compilation (Martin, Zalubas, & Hagan 1978). Unfortunately, the levels situated above  $40,000 \text{ cm}^{-1}$ are only partially known (for example, several levels belonging to  $6s^28d$ ,  $6s^29d$ ,  $6s^210d$  and 5d6s7s are missing); therefore some of the designations in the NIST compilation appear dubious. Moreover, some of these levels may overlap unknown levels belonging to higher configurations such as 5d6s6d, 5d6s7d,  $5d^3$ ,  $5d6p^2$ , .... Consequently, these levels above  $40,000 \text{ cm}^{-1}$  were not included in the fit. The possible effects of the  $6p^3$ ,  $6p^2 ns$  (n = 7-10),  $6p^2 np$  (n = 7-10) 10),  $6p^2$  nd (n = 6–10),  $6p^2$  nf (n = 5–10), 5d6p6d, and 6s6p6d were also investigated. It is worth noting that all possible subconfigurations and terms are automatically included for each configuration listed above in a calculation using Cowan's codes. This corresponds to more than 500 configurations in a fully relativistic or multiconfiguration Dirac-Fock calculation.

Five sets of calculations (A, B, C, D, and E) were performed in this work. In calculations A and B, the same ionic core was considered in the computations of the atomic orbitals and in the calculation of the polarization effects affecting the dipole operator. The only difference between calculations A and B consists of the use of different values of the cutoff radius  $r_c$ . Indeed, as already mentioned, for example, by Hibbert (1982b), this parameter is not unambiguously defined. In calculation A, we used  $r_c = 1.406 a_0$ , which corresponds to the expectation value of r for the outermost core orbitals  $(5p^6)$  as calculated by the Cowan's code. In the calculation B,  $r_c$  was chosen equal to 0.719  $a_0$ , which represents the distance at which the probability density of the core falls to 10% of its maximum value as suggested by Hameed (1972). Figure 1 shows the calculated probability density of the core in the ground configuration  $5d6s^2$  together with the  $r_c$  values used in the computations.



FIG. 1.—Electron probability density of the core in the ground configuration (5d6s<sup>2</sup>) of Lu I. The values of the cutoff radius used in calculations A ( $r_c = 1.406 a_0$ ) and B ( $r_c = 0.719 a_0$ ) are also represented on the figure.

In both calculations A and B, the value of the dipole polarizability,  $\alpha_d$ , was chosen to be equal to 5.20  $a_0^3$  as tabulated by Fraga, Karwowski, & Saxena (1976) for the Lu IV ion. In calculations C and D, the same core-polarization parameters ( $\alpha_d = 5.20 a_0^3$  and  $r_c = 1.406 a_0$ ) were used in the model potentials while different ionic cores were considered for the corrected dipole operators related to the different transition arrays. In the former case (C), a value of 28.27  $a_0^3$  for  $\alpha_d$  was chosen to correct for core polarization in the dipole matrix elements of all the transition arrays. It corresponds to a  $Lu^+$  core in the tables of Fraga et al. (1976). However, different cutoff radii,  $r_c$ , were used. They are equal to the geometric mean of the HFR average values  $\langle r \rangle$  for the outermost core orbitals of the corresponding configurations of Lu II when the active electron is removed in both states of the transition. In calculation D, different cores were considered for the initial and the final configurations of each transition array. More precisely, the dipole polarizabilities and the cutoff radii were obtained by considering the core as the remaining configuration when the active orbital is removed, the algebraic mean of  $\alpha_d$  values and the geometric mean of  $r_c$  values deduced, respectively, for the initial and the final configurations of a transition being used in the corrected dipole operator. As an example, if the transition array  $5d6s^2$ -5d6s6p is considered, the "core" configurations are 5d (Lu III-type:  $\alpha_d = 10.59 \ a_0^3$  and  $r_c(5d) = 2.54$  $a_0$ ) and 5d6s (Lu II-type:  $\alpha_d = 28.27 \ a_0^3$  and  $r_c(6s) = 3.44$  $a_0$ ) and the parameters used are  $\alpha_d = 19.43 a_0^3$  and  $r_c =$ 2.96  $a_0$ . Calculation E is similar to D, but we have added the configurations  $6p^3$ ,  $6p^2 ns (n = 7-10)$ ,  $6p^2 np (n = 7-10)$ ,  $6p^2$  nd (n = 6-10),  $6p^2$  nf (n = 5-10), 5d6p6d, and 6s6p6d in order to test their effect on the valence correlation.

## 5. DISCUSSION

Experimental and theoretical radiative lifetimes for Lu I from our work and from previous experiments are present-

ed in Table 1. Energy levels of Lu I are from Vergès & Wyart (1978). Our experimental results agree with the LIF measurements of Kwiatkowski, Teppner, & Zimmermann (1980) to within the combined uncertainties in all cases. Such good agreement between independent sets of LIF measurements has become common in recent years. The delayed-coincidence measurements electron-beam of Gorshkov, Komarovskii, & Penkin (1984) generally agree with our measurements for longer lifetimes but are in disagreement for many lifetimes shorter than about 20 ns, sometimes by more than 50%. As discussed in § 2, the fact that we can reproduce accurate, well-known lifetimes in our apparatus in the range of 1.8-8 ns makes such gross errors in our shorter lifetimes unlikely. The electron-beam used delayed-coincidence method by Gorshkov, Komarovskii, & Penkin (1984) does not provide the highly selective excitation of LIF methods. The older levelcrossing measurements by Göbel (1970a, 1970b, 1971) agree with our measurements to better than 15%.

Our theoretical HFR lifetime values (calculations A to E) are compared in Table 1 with our experimental lifetimes. The results of calculation D and E are expected to be the most accurate. For 15 levels, calculation D is in agreement with experiment within 30% while, for the remaining levels of the same table, the discrepancies are disappointingly large, at least when comparing with the situation encountered previously in Lu II (Quinet et al. 1999a). No systematic trend is observed when comparing theory with experiment, but a large scatter is present, thus indicating that an underor overestimation of the core-polarization effects must be ruled out. We observe that all the levels, for which theory and experiment disagree by more than 30%, are strongly perturbed (in fact, the leading percentages are in all cases smaller than 86%). In the case of calculation E, for half of the levels, the agreement with experiment is improved over that achieved when comparing calculation D to experiment while the agreement is somewhat deteriorated for the remaining half of the levels. In addition, in calculation E, some levels (22221.68, 24308.09, and 29486.94  $\text{cm}^{-1}$ ) appear particularly sensitive to cancellation effects in the calculation of the dominant depopulating transitions leading to inaccurate radiative lifetimes. The lifetimes obtained from calculation E for levels not affected by these cancellations are included in the last column of Table 1. The above discussion indicates that the large perturbations occurring in this complex neutral spectrum are not described with a sufficient accuracy by the physical model adopted in the calculations and that these effects are not adequately taken into account by the fitting procedure. Part of the discrepancies could also result from the fact that the highly excited levels are only partially known and that some adopted designations in the published analyses could be in need of revision. This discussion emphasizes the limits encountered in atomic structure calculations and particularly in the prediction of radiative atomic properties for a heavy system such as Lu I, where both complex configuration interaction and relativistic effects must be considered simultaneously.

Experimental and theoretical branching fractions for Lu I are presented in Table 4. The HFR calculations reveal that some of the Lu I upper levels contain significant branches in the infrared beyond the spectral limit of the FTS and the 1.0 m grating spectrometer. In some of these cases, we used the theoretical results to estimate the contribution of "missing" infrared branches to the total decay from an upper level and

adjust the other experimental results accordingly. These special cases are indicated by a superscript in Table 4. In two cases, namely, the branches from the  $6s^2 5f^2 F^o$  levels at 36633.31 and  $36644.12 \text{ cm}^{-1}$ , the HFR calculations alerted us to strong infrared transitions to the  $6s^26d^2D$  levels. Using the LIF apparatus, the infrared branching fraction from the  ${}^{2}F^{o}$  level was experimentally determined by comparing 288.5 nm fluorescence, resulting from direct decay of the  $6s^2 5f^2 F^o$  level at 36644.12 cm<sup>-1</sup>, with 412.5 nm fluorescence resulting from a cascade through the  $6s^26d \,^2D$  level at  $31713.60 \text{ cm}^{-1}$ . Filters were carefully chosen to block out all radiation but that from the transition of interest. The quantum efficiency of the fluorescence collection system, including the PMT, is approximately the same for 289 and 413 nm. Using the relative fluorescence intensities, the infrared branching fraction at 2027.6 nm from the  $6s^2 5f^2 F^o$  level was determined to be  $0.30 \pm 0.07$ . This result was used to correct the experimental branching fractions for the ultraviolet lines from the  $6s^25f \, {}^2F^o$  levels in Table 4. This approach of measuring cascade fluorescence was successful because of the very simple decay pattern of the  ${}^{2}F^{o}$  levels. In general, it is quite difficult to establish a reliable radiometric calibration over a large wavelength range from 280 nm to 2100 nm. Often several independent spectra must be pieced together to cover a decade of wavelength.

The agreement between the theoretical branching ratios (calculation D) and the experimental branching fractions is good for all the transitions except for the two transitions depopulating the level at 31523.03 cm<sup>-1</sup>, for which theory differs somewhat from experiment. Theory and experiment were carefully checked for that level. The only possible explanation of the observed difference probably is related to the fact that, within the configuration 5d6s6p, strong interactions occur between  ${}^{2}P^{o}$  and  ${}^{2}D^{o}$  levels. The theoretical branching fractions are extremely sensitive to the mixing between these two levels. If we exclude this particular case, Table 4 shows that the HFR method used here is adequate for providing reasonable estimates of the branching ratios of transitions for which such data cannot be determined experimentally. For most of the lines, the branching fractions of calculation E do not differ very much (a fraction of a 1% up to a few percent) from those of calculation D. In fact, for two-thirds of the lines, the results of calculation D are in closer agreement with experiment. For that reason, only those branching fractions are reported in Table 4. In addition, we observe that some of the transitions are strongly affected by cancellation effects that render the corresponding results unreliable.

Branching fractions are combined with radiative lifetimes to produce the Einstein A-coefficients and  $\log (gf)$  values presented in Table 5. A comparison between our experimentally determined f-values and f-values from older levelcrossing measurements is presented in Table 6. These are in agreement for two of the four cases. Transition wavenumber and wavelengths in Tables 5 and 6 were calculated directly from the energy levels (Vergès & Wyart 1978). These should be slightly more accurate than the wavenumbers and wavelengths of Table 4.

New measurements for the radiative lifetimes for Lu II are presented in Table 2 and are compared with previous experimental and theoretical results. Previously, the UW LIF apparatus was used to measure the radiative lifetime of the  $6s6p \, {}^{3}P^{o}_{1}$  level of Lu II at 28503.16 cm<sup>-1</sup> (Den Hartog et al. 1998). This single measurement was part of an effort to

TRANSITION						
Wavenumber Lambda in Air (cm <sup>-1</sup> ) (nm)		Upper Level Energy Parity $J$ (cm <sup>-1</sup> )	Lower Level Energy Parity $J$ (cm <sup>-1</sup> )	A-COEFFICIENT $(1.0E6 \text{ s}^{-1})$	log (gf)	
36633.36	272.894	36633.36 od 2.5	0.00 ev 1.5	$23.2 \pm 2.0$	-0.81	
34650.20	288.514	36644.12 od 3.5	1993.92 ev 2.5	$23.1~\pm~2.0$	-0.64	
34436.49	290.305	34436.49 od 1.5	0.00 ev 1.5	$7.9 \pm 0.5$	-1.40	
33443.10	298.928	33443.10 od 0.5	0.00 ev 1.5	$128 \pm 6$	-0.47	
32456.70	308.013	32456.70 od 2.5	0.00 ev 1.5	$7.2 \pm 0.6$	-1.21	
32442.57	308.148	34436.49 od 1.5	1993.92 ev 2.5	$262~\pm~14$	+0.17	
32058.10	311.843	32058.10 od 0.5	0.00 ev 1.5	$203~\pm~11$	-0.23	
31523.03	317.137	31523.03 od 1.5	0.00 ev 1.5	$66 \pm 3$	-0.40	
30488.62	327.897	30488.62 od 1.5	0.00 ev 1.5	$100 \pm 5$	-0.19	
30462.78	328.175	32456.70 od 2.5	1993.92 ev 2.5	$305 \pm 19$	+0.47	
30183.55	331.211	30183.55 od 2.5	0.00 ev 1.5	$185 \pm 9$	+0.26	
29757.25	335.956	31751.17 od 3.5	1993.92 ev 2.5	$244~\pm~12$	+0.52	
29608.01	337.649	29608.01 od 1.5	0.00 ev 1.5	168 ± 9	+0.06	
29529.11	338.552	31523.03 od 1.5	1993.92 ev 2.5	$50.3 \pm 2.7$	-0.46	
29431.05	339.680	29431.05 od 0.5	0.00 ev 1.5	$103 \pm 7$	-0.45	
28494.70	350.842	30488.62 od 1.5	1993.92 ev 2.5	$65 \pm 4$	-0.32	
28189.63	354.639	30183.55 od 2.5	1993.92 ev 2.5	$2.6 \pm 0.4$	-1.53	
28020.11	356.785	28020.11 od 2.5	0.00 ev 1.5	69 ± 4	-0.10	
27493.02	363.625	29486.94 od 3.5	1993.92 ev 2.5	6.4 ± 0.3	-0.99	
27406.11	364.778	31542.24 ev 1.5	4136.13 od 0.5	$92 \pm 5$	-0.13	
26026.19	384.119	28020.11 od 2.5	1993.92 ev 2.5	$26.5 \pm 1.6$	-0.45	
25191.56	396.846	25191.56 od 2.5	0.00 ev 1.5	$2.21 \pm 0.14$	-1.51	
24657.29	405.445	28793.42 ev 0.5	4136.13 od 0.5	$20.1 \pm 1.1$	-1.00	
24308.09	411.270	24308.09 od 1.5	0.00 ev 1.5	$0.43 \pm 0.06$	-2.36	
24237.21	412.472	31713.60 ev 2.5	7476.39 od 1.5	$\stackrel{-}{89 \pm 9}$	+0.14	
24065.85	415.409	31542.24 ev 1.5	7476.39 od 1.5	16.8 + 1.1	-0.76	
23197.64	430.957	25191.56 od 2.5	1993.92 ev 2.5	$0.66 \pm 0.04$	-1.96	
22314.17	448.020	24308.09 od 1.5	1993.92 ev 2.5	$0.088 \pm 0.012$	-2.97	
22221.68	449.885	22221.68 od 2.5	0.00 ev 1.5	$0.128 \pm 0.009$	-2.63	
22124.76	451.856	22124.76 od 1.5	0.00 ev 1.5	22.6 + 1.2	-0.56	
21462.38	465.801	21462.38 od 2.5	0.00 ev 1.5	3.15 + 0.18	-1.21	
21317.03	468.977	28793.42 ev 0.5	7476.39 od 1.5	1.32 + 0.14	-2.06	
21195.37	471.669	21195.37 od 1.5	0.00 ev 1.5	0.113 + 0.012	$-2.82^{a}$	
20762.47	481.504	20762.47 od 0.5	0.00 ev 1.5	0.90 + 0.06	-2.20	
20227.76	494.232	22221.68 od 2.5	1993.92 ev 2.5	0.97 + 0.06	-1.67	
20130.84	496.612	22124.76 od 1.5	1993.92 ev 2.5	0.033 + 0.005	-3.31	
19989.86	500.114	24125.99 ev 0.5	4136.13 od 0.5	32.0 + 1.8	-0.62	
19468.46	513.508	21462.38 od 2.5	1993.92 ev 2.5	9.1 + 0.5	-0.67	
19201.45	520.649	21195.37 od 1.5	1993.92 ev 2.5	$0.254 \pm 0.026$	-2.38ª	
18504.58	540.257	18504.58 od 2.5	0.00 ev 1.5	1.20 + 0.06	-1.50	
17427.28	573.654	17427.28 od 1.5	0.00 ev 1.5	1.73 + 0.09	-1.47	
16649.60	600.449	24125.99 ev 0.5	7476.39 od 1.5	49.3 + 2.6	-0.27	
16510.66	605.502	18504.58 od 2.5	1993.92 ev 2.5	0.92 + 0.05	-1.52	
15433.36		17427.28 od 1.5	1993.92 ev 2.5	$0.072 \pm 0.006$	-2.74	

TABLE 5 ATOMIC TRANSITION PROBABILITIES FOR LU I

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<sup>a</sup> Transition probabilities from the  ${}^{4}D$  level at 21195.37 cm<sup>-1</sup> are based on a lifetime from Kwiatkowski et al. 1980.

TABLE 6
Comparison of Experimental $f$ -Values for Lu i

TRANSITION				Absorption <i>f</i> -value		
Wavenumber (cm <sup>-1</sup> )	Lambda in Air (nm)	Upper-Level Energy Parity $J$ (cm <sup>-1</sup> )	Lower-Level Energy Parity $J$ (cm $^{-1}$ )	This Experiment	Other Experiment	
29608.01 28020.11 26026.19 22124.76	337.649 356.785 384.119 451.856	29608.01 od 1.5 28020.11 od 2.5 28020.11 od 2.5 22124.76 od 1.5	0.00 ev 1.5 0.00 ev 1.5 1993.92 ev 2.5 0.00 ev 1.5	$\begin{array}{c} 0.287 \ \pm \ 0.015 \\ 0.197 \ \pm \ 0.010 \\ 0.059 \ \pm \ 0.003 \\ 0.069 \ \pm \ 0.004 \end{array}$	$\begin{array}{c} 0.382  \pm  0.015^a \\ 0.170  \pm  0.020^b \\ 0.056  \pm  0.008^b \\ 0.063  \pm  0.001^b \end{array}$	

<sup>a</sup> Göbel 1971. <sup>b</sup> Göbel 1970b.

refine the solar abundance of lutetium. Early solar abundance estimates were in error because the solar line at 339.7062 nm used to determine the lutetium abundance was presumed to be caused solely by Lu II (Moore, Minnaert, & Houtgast 1966) but has subsequently been found to be blended with NH (Bord, Cowley, & Mirijanian 1998). The nearly unblended solar line at 622.187 nm has been used recently to determine a more reliable solar lutetium abundance (Bord et al. 1998; Den Hartog et al. 1998). Accurate experimentally determined radiative lifetimes for Lu II have also been measured by using LIF from a laser-produced plasma followed by time-resolved detection (Li & Lundberg 1998) and by using the fast-beam-laser method (Quinet et al. 1999a). Some radiative lifetimes for Lu II have been measured using a beam-foil technique (Andersen & Sorensen 1974; Andersen et al. 1975). Our new experimental results agree well with the recent measurements of Li & Lundberg (1998) and agree with three of the four measurements using the fast-beam-laser reported in Quinet et al. (1999a). It has been suggested that the error bars on the level at 48773.19  $\text{cm}^{-1}$  (and perhaps at 44918.68  $\text{cm}^{-1}$ ) are too small (E. H. Pinnington, 1999, private communication). In general, for short lifetimes (< 20 ns), the fast-beam-laser method used by the Edmonton group is more accurate than the timeresolved LIF method used in the work reported here. The difficulty on the Lu II levels at 48733.18 cm<sup>-1</sup> (and perhaps at 44918.68  $\text{cm}^{-1}$ ) appears to have been caused by poor signal-to-noise ratios in the fast-beam-laser experiment (E. H. Pinnington, 1999, private communication). It is our opinion that most of the earlier lifetime measurements by the Edmonton group, especially those on the iron group elements, are still the best available. We argue that the new measurements reported here for these two Lu II levels should be more accurate than the fast-beam-laser measurements reported in Quinet et al. (1999a). The fast-beam-laser method has been used to measure lifetimes to 1% or better accuracy because systematic effects can be well understood and controlled. The time-resolved LIF method used here is less suited to measurements of 1% uncertainty because of difficulties in understanding and controlling systematic errors from the bandwidth, linearity, and overall fidelity of the electronic detection system at the 1% level. However, the UW time-resolved LIF experiment has a major advantage in high signal levels and high data collection rates, which enables us to make large sets of measurements with uncertainties of 5%. The beam foil measurements of Ander-

- Adams, D. L., & Whaling, W. 1981, J. Opt. Soc. Am., 71, 1036 Andersen, T., Poulsen, O., Ramunujam, P. S., & Petrakiev Petkov, A. 1975, Sol. Phys., 44, 257
- Andersen, T., & Sorensen, G. 1974, Sol. Phys., 38, 343
- Biémont, E., Baudoux, M., Kurucz, R. L., Ansbacher, W., & Pinnington, E. H. 1991, A&A, 249, 539
- Biémont, E., Dutrieux, J.-F., Martin, I., & Quinet, P. 1998a, J. Phys. B, 31, 3321
- Biémont, E., Li, Z. S., Palmeri, P., & Quinet, P. 1999, J. Phys. B, 32, 3409
- Biémont, E., Quinet, P., & Van Renterghem, V. 1998b, J. Phys. B, 31, 5301
- Bord, D. J., Cowley, C. R., & Mirijanian, D. 1998, Sol. Phys., 178, 221 Bridges, J. M., & Ott, W. R. 1977, Appl. Opt., 16, 367

- Carlsson, J., Sturesson, L., & Svanberg, S. 1989, Z. Phys. D, 11, 287 Cowan, J. J., Sneden, C., Truran, J. W., & Burris, D. L. 1996, ApJ, 460, L115
- Cowan, R. D. 1981, The Theory of Atomic Structure and Spectra (Berkeley: Univ. California Press)
- Cowley, C. R., & Mathys, G. 1998, Á&A, 339, 165
- Curry, J. J., Den Hartog, E. A., & Lawler, J. E. 1997, J. Opt. Soc. Am. B, 14, 2788

sen et al. (1975) and Andersen & Sorensen (1974) agree with our measurements in a few cases. Cascade repopulation may have been a problem in the beam foil experiments on Lu II levels at 48733.18 and 50049.20 cm<sup>-1</sup>, where the beam foil results are longer than our lifetime measurements. Line blending may have been a problem in the beam foil experiments on Lu II levels at 41224.96, 44918.68, and 45532.33 cm<sup>-1</sup>, where the beam foil results are shorter than our lifetime measurements. Except for the three longest lifetimes, the previous HFR calculations of Quinet et al. (1999a) are systematically about 10% lower than the measurements reported here; otherwise, the agreement is quite good.

Radiative lifetime for two odd-parity levels of Lu III at 44705.21 and 38400.61 cm<sup>-1</sup> were also measured; these are presented in Table 3. The lifetime of the 44705.21 cm<sup>-1</sup> level is too short for the electronic bandwidth of our apparatus, but we determined an upper bound of  $\tau < 1.8$  ns for this level. We are unaware of other experimental work on Lu III, but our measurements agree well with calculated lifetimes by Biémont et al. (1999) and Migdalek (1982).

The present results, along with results for Lu II published by Quinet et al. (1999a), provide an extensive set of radiative lifetimes, branching fractions, and f-values for Lu I and Lu II. Although substantial progress has been made in HFR calculations of *f*-values in rare earth spectra, more work will be required before theoretical f-values can replace highquality experimental f-values for rare earth spectra with more than three-valence electrons. However, the accuracy of branching fractions from the HFR calculations is quite encouraging. Branching fractions for lines widely separated in wavelength, such as those from the  $6s^25f^2F^o$  of Lu I, are very difficult to measure because of the necessity of establishing an accurate radiometric calibration over a large range of wavelengths. Studies of "r" versus "s" nucleosynthesis of heavy elements and the needs of the lighting research community provide strong motivation to continue improving both the experiments and the calculations.

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REFERENCES

- Danzmann, K., & Kock, M. 1982, J. Opt. Soc. Am., 72, 1556 Den Hartog, E. A., Curry, J. J., Wickliffe, M. E., & Lawler, J. E. 1998, Sol. Phys., 178, 239
- Den Hartog, E. A., Wiese, L. M., & Lawler, J. E. 1999, J. Opt. Soc. Am. B, 16, 2278
- Fraga, S., Karwowski, J., & Saxena, K. M. S. 1976, Handbook of Atomic Data (Amsterdam: Elsevier)
- Göbel, L. H. 1970a, Z. Naturforsch. A, 25, 1401
- . 1970b, Z. Naturforsch. A, 25, 611 1971, Z. Naturforsch. A, 26, 1559
- Gorshkov, V. N., Komarovskii, V. A., & Penkin, N. P. 1984, Opt. Spectrosc. (USSR), 56, 575 (original in Opt. Spektrosk., 56, 939) Guo, B., Ansbacher, W., Pinnington, E. H., Ji, Q., & Berends, R. W. 1992,
- Phys. Rev. A, 46, 641 Hameed, S. 1972, J. Phys. B, 5, 746
- Hashiguchi, S., & Hasikuni, M. 1985, J. Phys. Soc. Japan, 54, 1290
- Hibbert, A. 1982a, Adv. At. Mol. Phys. 18, 309
- 1982b, Nucl. Instrum. Methods Phys. Res., 202, 323
- Kono, A., & Hattori, S. 1984, Phys. Rev. A, 29, 2981
- Kwiatkowski, M., Teppner, U., & Zimmermann, P. 1980, Z. Naturforsch. A, 35, 370

- Lawler J. E. 1988, in Lasers, Spectroscopy, and New Ideas: A Tribute to Arthur L. Schawlow, ed. W. M. Yen & M. D. Levenson (New York: Springer), 125
- Li, Z., & Lundberg, H. 1998, in Abstracts of Contributed Oral Papers and Poster Papers from the Sixth Int. Collog. on Atomic Spectra and Oscil-lator Strengths for Astrophysical and Laboratory Plasmas, ed. J. Tatum

- lator Strengths for Astrophysical and Laboratory Plasmas, ed. J. Tatum (Victoria, B C, Canada: Univ. Victoria), 98
  Marsden, G. C., Den Hartog, E. A., Lawler, J. E., Dakin, J. T., & Roberts, V. D. 1988, J. Opt. Soc. Am. B, 5, 606
  Martin, W. C., Zalubas, R., & Hagan, L. 1978, Atomic Energy Levels: The Rare Earth Elements (NSRDS-NBS 60) (Washington: GPO)
  Meggers, W. F., Corliss, C. H., & Scribner, B. F. 1975, NBS Monograph 145, part I, Tables of Spectral Line Intensities (Washington: GPO), 136
  Migdalek, J. 1982, J. Quant. Spectrosc. Radiat. Transfer, 28, 417
  Migdalek, J., & Baylis, W. E. 1978, J. Phys. B, 11, L497
  Moore, C. E., Minnaert, M. G. J., & Houtgast, J. 1966, NBS Monograph 61, The Star Spectrum, 2935 Å to 8770 Å (Washington: GPO)
  Nitz, D. E., Bergeson, S. D., & Lawler, J. E. 1995, J. Opt. Soc. Am. B, 12, 377

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- Palmeri, P., Quinet, P., Frémat Y., Wyart, J.-F., & Biémont, E. 2000a, ApJS, 129, 367
- Palmeri, P., Quinet, P., Wyart J.-F., & Biémont, E. 2000b, Phys. Scr., 61, 323
- Quinet, P., Palmeri, P., & Biémont, E. 1999b, J. Quant. Spectrosc. Radiat. Transfer, 62, 625
- Transfer, 62, 625
  Quinet, P., Palmeri, P., Biémont, E., McCurdy, M. M., Rieger, G., Pinnington, E. H., Wickliffe, M. E., & Lawler, J. E. 1999a, MNRAS, 307, 934
  Smith, V. V., Cunha, K., & Lambert, D. L. 1995, AJ, 110, 2827
  Sneden, C., Basri, G., Boesgaard, A. M., Brown, J. A., Carney, B. W., Kraft, R. P., Smith, V., & Suntzeff, N. B. 1995, PASP, 107, 997
  Sneden, C., McWilliam, G. W., Preston, G. W., Cowan, J. J., Burris, D. L., & Armosky, B. J. 1996, ApJ, 467, 819
  Vergès, J., & Wyart, J. F. 1978, Phys. Scr., 17, 495
  Whaling, W., Carle, M. T., & Pitt, M. L. 1993, J. Quant. Spectrosc. Radiat. Transfer, 50, 7
  Weiss, A. W. 1995, Phys. Rev. A, 51, 1067
  Woolf, V. M., Tomkin, J., & Lambert, D. L. 1995, ApJ, 453, 660