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*Published in:*  
Optical Society of America. Journal B: Optical Physics

2006

[Link to publication](#)

*Citation for published version (APA):*

Xu, H., Jiang, Z., & Lundberg, H. (2006). Lifetime measurements in neutral and singly ionized vanadium. *Optical Society of America. Journal B: Optical Physics*, 23(12), 2597-2600.  
<http://www.opticsinfobase.org/abstract.cfm?URI=josab-23-12-2597>

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3

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# Lifetime measurements in neutral and singly ionized vanadium

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Received August 4, 2006; revised August 24, 2006; accepted August 27, 2006; posted September 1, 2006 (Doc. ID 73796)

Radiative lifetimes of ten odd-parity levels of V I belonging to the  $3d^34s4p$  and  $3d^44p$  configurations and of 11 odd-parity levels of V II belonging to the  $3d^34p$  configuration are reported. The lifetimes were measured with use of single-step excitation and time-resolved fluorescence spectroscopy. The neutral and singly ionized vanadium atoms were produced in a laser-induced vanadium plasma. © 2006 Optical Society of America  
OCIS codes: 300.2530, 020.4900, 300.6500.

## 1. INTRODUCTION

Radiative lifetimes are fundamental radiative properties of atoms and ions and can be used for obtaining oscillator strengths when they are combined with intensity measurements (branching ratios).<sup>1</sup> Oscillator strengths are of great importance for determining the abundance of elements in solar and stellar photospheres<sup>2</sup> and also have applications in other fields, such as plasma physics and laser chemistry. In recent years, a lot of work has been performed with regard to radiative lifetimes for neutral and singly ionized elements of the iron group.<sup>3,4</sup> Vanadium belongs to this group ( $Z=23$ ). In astrophysics, the V I and V II spectral lines have been observed in the solar photosphere and many stars.<sup>5–8</sup> The previous measurements of radiative lifetimes and transition probabilities undertaken in V I and V II were focused mainly on low-lying excited states, and the data for high-lying excited states of V I and V II are lacking.<sup>7,9–14</sup> To get more insight into the radiative properties of vanadium, additional lifetime measurements are required.

A large number of methods, such as the beam-foil technique, the hook method, and the delayed-coincidence techniques following electron-beam excitation, have been developed for measuring radiative lifetimes of excited atomic or ionic levels. Among the methods available, time-resolved laser-induced fluorescence (LIF) techniques based on selective laser excitation of the required level provide direct means of obtaining lifetimes and have proven to be very reliable for radiative lifetime measurements on neutral and singly ionized atoms, as well as on doubly ionized atoms. A lot of work has been undertaken using this technique applied to different atomization

sources, for example, hollow cathodes, thermal ovens, and laser-induced plasmas.

In the present work, we performed lifetime measurements on neutral and singly ionized vanadium atoms by the method of time-resolved LIF in a laser-induced plasma. Ten odd-parity levels of V I with energy from 30 635 to 41 930  $\text{cm}^{-1}$  and 11 odd-parity levels of V II with energy from 39 400 to 47 420  $\text{cm}^{-1}$  are reported. In the measurements, frequency upconversion in KDP and  $\beta$ -barium borate (BBO) crystals made the measurements of lifetimes in the short-wavelength spectral range feasible. Also, the methods of laser compression by stimulated Brillouin scattering (SBS) in water<sup>15</sup> and laser-wavelength extension by stimulated Stokes Raman scattering (SSRS) in hydrogen gas were used to obtain a short excitation laser pulse at suitable excitation wavelength.

## 2. EXPERIMENTAL SETUP

The experimental setup used for the lifetime measurements has been described in earlier papers.<sup>15</sup> A pure vanadium foil was used as a rotating target in a vacuum chamber, in which the background pressure was about  $10^{-6}$ – $10^{-5}$  mbar. Free neutral and singly ionized vanadium atoms were produced by laser ablation, induced with a 532 nm laser pulse emitted from a Nd:YAG laser (Continuum Surelite). The pulse length of the ablation pulse was 10 ns, and the repetition rate was 10 Hz. The pulse energy was usually in the range of 2–10 mJ. The ablation pulse was directed from the top of the vacuum system through a glass window and was vertically fo-

cused onto the vanadium foil. After the impinging of the laser pulse on the vanadium foil, a small plasma containing electrons, atoms, and ions of various ionization stages of vanadium was produced. With appropriately chosen plasma conditions, sufficient population in ground as well as metastable states of the neutral and singly ionized vanadium was obtained.

To obtain the required excitation radiation, we employed a pulsed 532 nm laser beam from another Nd:YAG laser (Continuum NY-82), characterized by 8 ns pulse duration, single-pulse energy of 400 mJ, and 10 Hz repetition rate. The pulses were first passed through a SBS system, similar to the one described by Schiemann *et al.*<sup>16,17</sup> The methods of laser-pulse compression by SBS in water can result in a pulse output with about 1 ns duration and about 150 mJ single-pulse energy. For obtaining a tunable excitation source, a dye laser (Continuum Nd-60) was employed and pumped by the temporarily compressed laser pulses. 4-dicyanomethylene-2-methyl-6-(*p*-dimethylaminostyryl)-4*H*-pyran (DCM) dye was used in the experiments. The dye laser output was frequency doubled in a KDP crystal or frequency tripled in a system consisting of a KDP crystal, a retarding plate, and a BBO crystal. The first- or second-order Stokes component (*S*) of Raman shifting was obtained when the second harmonic ( $2\omega$ ) or the third harmonic ( $3\omega$ ) of the dye laser radiation was focused into a SSRS cell with hydrogen gas at a pressure of 10 bars. The appropriate radiation component from the SSRS cell was selected using a Pellin–Broca prism and was then horizontally sent into the vacuum chamber about 10 mm above the vanadium foil. Both Nd:YAG lasers were externally triggered by a delay generator.

After the interaction between the excitation pulse and the plasma, the released fluorescence from the excited levels was collected by a fused-silica lens and then appropriately filtered by a 1/8 m monochromator, finally to be detected by a Hamamatsu 1564U microchannel-plate photomultiplier tube with a rise time of 0.2 ns. The time-resolved signal from the detector was captured by a transient recorder (Tektronix Model DSA 602), which has a 1 GHz bandwidth and a 2 G samples/s real-time sampling rate. The decay data were transferred to a personal computer for lifetime evaluation.

### 3. MEASUREMENTS AND RESULTS

The ground electronic configurations of the V I and V II are  $3d^34s^2$  and  $3d^4$ , respectively. Through single-step excitation from the ground state and appropriate metastable states, radiative lifetimes of ten odd-parity levels of V I belonging to the  $3d^34s4p$  and  $3d^44p$  configurations and of 11 odd-parity levels of V II belonging to the  $3d^34p$  configuration were measured. The energy levels in the experiment were taken from the National Institute of Standards and Technology atomic spectra database ([http://www.physics.nist.gov/cgi-bin/AtData/main\\_asd](http://www.physics.nist.gov/cgi-bin/AtData/main_asd)). The excitation lines for the studied levels are in the region 210–330 nm. The region can be covered by different nonlinear processes ( $2\omega$ ,  $3\omega+S$ ,  $3\omega+2S$ , and  $3\omega$ ) using the DCM dye operated in the wavelength region 629–658 nm. Fluorescence, released at the decay of the investigated

levels to possible lower levels, was checked using the monochromator, and the strongest transition was usually used for the recordings. The levels measured are summarized in Table 1.

Possible systematic errors were carefully investigated in this experiment. Attempts were made to observe lifetime variations by changing various experimental parameters. The delay times between the ablation and the excitation pulses were varied and could be as long as 8  $\mu$ s for the V I measurements and 4  $\mu$ s for V II, while a sufficient signal-to-noise ratio for evaluating the lifetime was maintained. In this way, the detected fluorescence intensity was varied by a factor of 10, but the lifetime values were found to be well coincident. This indicates that radiation trapping and collisional quenching effects were absent for the experimental conditions selected.<sup>18</sup> The difference in achievable delay times between V I and V II is due to the lower speeds of atoms compared with ions. An important aspect in lifetime measurements is to avoid the flight-out-of-view effects, especially when the measured lifetimes are long. In our experiment, three radiative lifetimes of the  $3d^34s4p\ ^4G^0$  terms of neutral vanadium atoms are longer than 70 ns. Flight-out-of-view effects for these three lifetimes were, besides using long delay times, minimized by adjusting the position of the monochromator entrance slit and opening it maximally (3 mm).

In this experiment, we adopted different fitting processes for the evaluation of lifetimes. For the lifetimes of the  $3d^34s4p\ ^4G^0$  terms in V I that were longer than 10 ns, a least-squares exponential fitting procedure was applied to the decay at times when the excitation pulse had vanished. On the other hand, for other lifetimes in V I and V II that were shorter than 10 ns, a deconvolution procedure was performed. In this fitting procedure, the temporal shape of the excitation pulses was recorded by inserting a metal rod into the interaction zone of the excitation laser and the plasma when the ablation beam was blocked. We performed the evaluation process by fitting the experimental fluorescence decay curve to a convolution of the recorded excitation pulse and a pure exponential function.<sup>19</sup> Typical experimental curves for the exponential (V I: 30 635.59  $\text{cm}^{-1}$ ) and the deconvolution fitting procedures (V II: 46 690.42  $\text{cm}^{-1}$ ) are shown in Fig. 1.

To obtain a smooth decay curve with a reasonable signal-to-noise ratio in the measurements, we obtained the decay curve of each level to be investigated by averaging fluorescence photons from 1000 pulses. About ten curves under different experimental conditions were recorded for each level investigated, and the final lifetime value was taken as the average of the data from these curves. The uncertainties reported in Table 1 include the statistical uncertainties.

### 4. DISCUSSION

The previous lifetime measurements in V I and V II were primarily concentrated on the low-lying levels. Only the beam-foil measurements by Roberts *et al.* present several experimental lifetimes of V II belonging to high-lying levels.<sup>9</sup> However, the discrepancies between the values obtained by Roberts *et al.* and those obtained by the later

**Table 1. Radiative Lifetimes Measured for V I and V II**

Configuration	Term	J	Energy (cm <sup>-1</sup> )	Excitation		Observed λ (nm) <sub>vac.</sub>	Lifetime (ns)	
				Origin	λ (nm) <sub>vac.</sub>		This Work	Previous
V I								
3d <sup>3</sup> 4s4p	y <sup>4</sup> G <sup>0</sup>	2.5	30 635.59	0.0	326.417	326	74 (5)	
		3.5	30 694.35	137.38	327.257	327	87 (6)	
		4.5	30 771.73	323.46	328.425	328	105 (10)	
3d <sup>4</sup> 4p	t <sup>4</sup> D <sup>0</sup>	0.5	37 757.30	0.0	264.849	354	5.3 (0.4)	
		3.5	38 115.69	323.46	264.604	353	5.6 (0.4)	
		4.5	39 391.08	323.46	255.966	423	9.6 (0.7)	
	u <sup>4</sup> F <sup>0</sup>	1.5	39 266.68	0.0	254.668	424	8.7 (0.6)	
		2.5	39 300.56	137.38	255.341	424	9.4 (0.7)	
		3.5	39 341.79	323.46	256.289	423	9.5 (0.7)	
3d <sup>3</sup> 4s4p	r <sup>4</sup> D <sup>0</sup>	0.5	41 928.48	0.0	238.501	309	7.3 (0.5)	
V II								
3d <sup>3</sup> 4p	z <sup>3</sup> G <sup>0</sup>	4	39 403.74	208.89	255.135	327	4.7 (0.3)	7.4 <sup>a</sup> 4.8 <sup>b</sup>
		5	39 612.96	339.21	254.622	328	4.7 (0.3)	6.9 <sup>a</sup> 4.7 <sup>b</sup>
	y <sup>5</sup> D <sup>0</sup>	1	46 690.42	36.05	214.342	301	5.5 (0.4)	
		2	47 101.89	36.05	212.468	298	5.0 (0.4)	6.5 <sup>a</sup>
		3	47 181.21	106.63	212.428	299	4.6 (0.3)	5.5 <sup>a</sup>
		4	47 420.25	208.89	211.813	297	4.7 (0.3)	
	z <sup>3</sup> P <sup>0</sup>	2	46 739.99	106.63	214.438	301	6.0 (0.5)	9.9 <sup>a</sup>
		1	47 107.99	106.63	212.759	298	5.1 (0.4)	
	z <sup>5</sup> P <sup>0</sup>	1	46 754.59	0.0	213.882	302	2.9 (0.3)	
		2	46 879.98	36.05	213.474	302	2.7 (0.3)	5.2 <sup>a</sup>
		3	47 051.86	106.63	213.014	300	2.7 (0.3)	

<sup>a</sup>Roberts *et al.*<sup>9</sup>

<sup>b</sup>Biémont *et al.*<sup>14</sup>

measurements by Biémont *et al.* for the low-lying levels were large.<sup>14</sup> Therefore, the lifetime data from the present investigations are compared in Table 1 with the previously published results. Two levels of V II earlier measured by Biémont *et al.* using the time-resolved LIF techniques were found to be in good agreement with our results. But six lifetimes obtained for V II are much smaller than those reported by Roberts *et al.* The discrepancies are not uncommon because of the possibilities of cascade repopulation due to nonselective excitation in the beam-foil measurements.

In summary, radiative lifetimes of ten odd-parity levels for V I and 11 odd-parity levels for V II have been obtained using time-resolved LIF techniques combined with the methods of laser-pulse compression by SBS in water, laser-wavelength extension by SSRS in hydrogen gas, and frequency upconversion in crystals. The results presented here are expected to provide the astrophysicists with reliable radiative data needed for investigating chemical composition of stellar objects.

**ACKNOWLEDGMENTS**

This work was financially supported by the Swedish Research Council and by the National Natural Science Foundation of China (10274025). We thank Z. Li for participating in the early part of the measurements and A. Persson for technical help. The support by S. Svanberg is gratefully acknowledged.

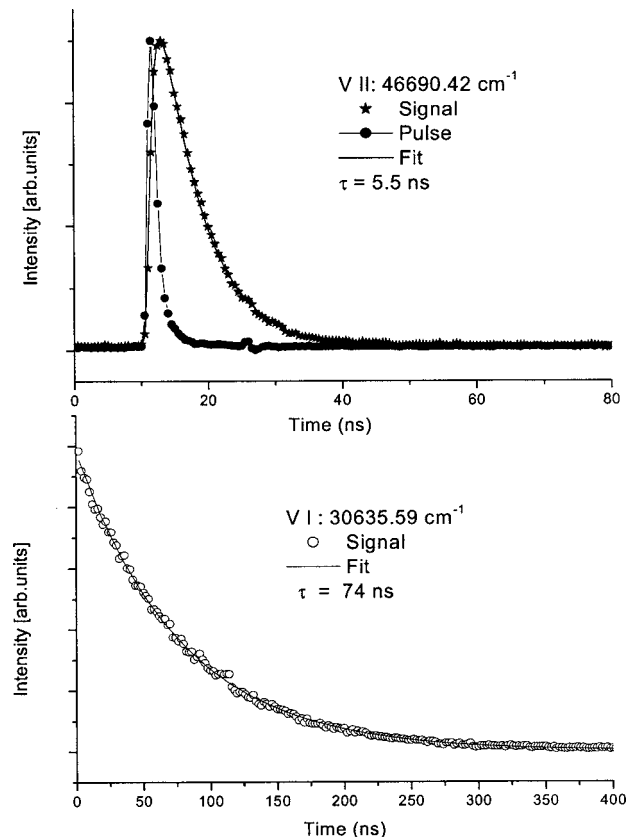


Fig. 1. Decay curves recorded for the 46 690.42 cm<sup>-1</sup> level of V II and the 30 635.59 cm<sup>-1</sup> level of V I.

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