Pulsed ion source for laser spectroscopy: Application to Nb II

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A novel pulsed ion source suitable for laser spectroscopy on any metallic element is described. The source is based on a large-bore low-pressure hollow-cathode discharge. The first direct radiative lifetime measurements in Nb II are reported. Radiative lifetimes for z^5G and z^3D levels of Nb II are determined using time-resolved laser-induced fluorescence on a Nb II ion beam.

We describe a pulsed ion source suitable for laser spectroscopy, and an application of this source in the first direct radiative lifetime measurements on singly ionized niobium (Nb II). The pulsed ion source we developed is based on a large-bore, low-pressure hollow-cathode discharge. Duquette, Salih, and Lawler used a steady-state hollow-cathode discharge to make atomic beams of Mo, W, Zr, Nb, Hf, Re, Rh, Ta, and other elements for radiative lifetime measurements.[1,2] The hollow-cathode discharge provides a simple, very reliable method of producing free atoms of any metallic element. We discovered that a hollow cathode can also be used as an ion source by pulsing the discharge with a 50-kW power supply. The source is capable of producing ions of any metallic element in a variety of metastable levels.

Radiative lifetimes determined using time-resolved laser-induced fluorescence on an atomic beam or ion beam are very reliable ($\sim 5\%$). Laser excitation is selective; it greatly reduces problems due to cascades. The beam environment greatly reduces problems due to collisional quenching and radiation trapping. The radiative lifetimes are combined with branching ratios to produce accurate oscillator strengths (f values).

There is considerable interest in measuring accurate f values of refractory atoms and ions. For example, improved f values in Rh I resolved the discrepancies between the solar abundance of Rh determined spectroscopically and the solar system abundance determined from chondritic meteorites.[3,4] The solar abundance problem provides special motivation for improving f values in Nb I and Nb II. Radiative lifetimes in Nb I were measured using time-resolved laser-induced fluorescence by Duquette and Lawler,[2] Rudolph and Helbig,[5] and Kwiatkowski, Zimmerman, Biemont, and Grevesse.[3] All three independent sets of measurements are in excellent agreement, and improved Nb I f values have resulted from these measurements. There remains a substantial disagreement between the solar abundance of Nb and the abundance of Nb determined from chondritic meteorites. Spectral lines of Nb II have been identified in the solar spectrum.[6] The only f values available in Nb II are from the extensive emission measurements of Corliss and Bozman (CB).[7] The lifetime measurements of Duquette and Lawler indicate that the CB f values for Nb I have a substantial energy-dependent error.[2] The CB f values for Nb II, therefore, need to be checked. Our lifetime measurements in Nb II provide this check.

Figure 1 is a schematic of the experiment. Our apparatus has features in common with the apparatus used by Duquette, Salih, and Lawler to produce atomic beams.[1,2] Reference 2 contains a detailed description of the atomic beam apparatus including a drawing of the source showing materials and dimensions. The beam source is based on a low-pressure, large-bore hollow-cathode discharge. The hollow cathode is used as a beam source by sealing one end of the cathode except for a 1.0-mm-diam opening. The opening is flared outward at 45° to serve as a nozzle for forming an uncollimated ion beam. The hollow cathode and the scattering chamber are at ground potential. Argon, the sputtering gas, is continuously fed into the hollow-cathode discharge. A 6-in. diffusion pump is used to pump the scattering chamber. It is sealed from the hollow-cathode discharge except for the nozzle and is maintained at a much lower pressure than the discharge. The argon pressure in the discharge is typically 0.3 Torr. The resulting scattering chamber pressure is approximately 1.0×10^{-4} Torr.

The most significant change from the experiments on neutral atoms is the addition of the pulsed power supply. The pulsed supply is constructed of a storage capacitor, a triggerable switch, and a pulsed modulator transformer. The storage capacitor (0.85 μF) is charged through a current limiting inductor (15 H). The inductor allows the switch to recover between pulses at repetition rates up to 200 Hz. The switch is a silicon controlled rectifier (Motorola SCR 1718-8). The silicon controlled rectifier is rated for a peak reverse blocking voltage of 600 V and a peak forward

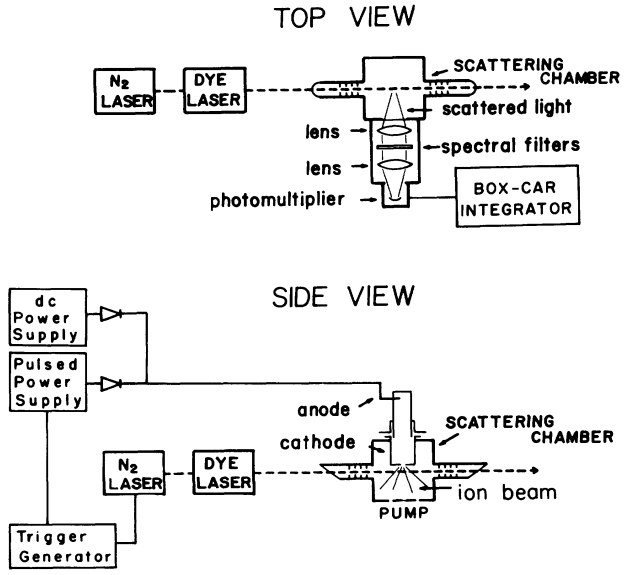

FIG. 1. Schematic of the experiment.

surge current of 1000 A. A surplus radar transformer steps up the voltage by a factor of 7. The supply is easily capable of driving a 25-A current pulse of 4-μsec duration into the hollow-cathode discharge. The discharge current is maintained at approximately 100 mA between pulses by a separate dc supply.

The ion beam is crossed by a pulsed dye laser beam 1 cm from the nozzle. The dye laser is pumped by a pulsed N_2 laser. The N_2 laser produces a 0.5-MW pulse of 337.1-nm radiation of 4-nsec full width at half maximum (FWHM) duration. The dye laser produces a pulse of 3-nsec duration (FWHM) with a 0.2-cm^{-1} bandwidth and with a peak power of up to 50 kW. A potassium dihydrogen phosphate (KDP) crystal frequency doubler extends the dye laser tuning range to 260 nm in the ultraviolet. The fluorescence is detected along an axis orthogonal to the atomic beam and laser beam. In order to minimize scattered light, several sets of light baffles are arranged along the laser beam axis inside of the Brewster windows that pass the laser beam into and out of the scattering chamber. Fluorescence from the scattering chamber is focused on the photomultiplier by two lenses comprising an $f/1$ system. The bias resistors of the 1P28A photomultiplier are bypassed by capacitors to insure good linearity at large peak currents. All components are wired for very low overall inductance and fast response. Signals from the photomultiplier pass through a delay line for synchronization of electronic components, then to a Princeton applied research model 162/163/165 box-car averaging system. Curves are plotted directly on an X-Y recorder.

The experiment includes a trigger generator to provide an adjustable delay between the current pulse to the hollow-cathode discharge and the laser pulse. The delay is measured from the peak of the discharge current pulse to the laser pulse. The delay which produces the largest ion fluorescence signal varies from element to element. A 12.5-μsec delay is optimum for Hf II, a 20-μsec delay is optimum for Nb II, and a 8.5-μsec delay is optimum for Fe II. The fluorescence excitation and observation region is 1 cm from the source nozzle. The optimum delay provides an indication of the average ion velocity.

We verified that the delay between the peak of the discharge current and the optimum laser-induced fluorescence signal is due to time of flight by studying Doppler shifts and broadenings of an ion line. A Doppler shift is detected by comparing a fluorescence signal observed with the laser propagating parallel to the ion beam axis to a fluorescence signal observed with the laser propagating perpendicular to the ion beam axis. The average ion velocity is important because it sets an upper limit for lifetimes which can be measured in the apparatus due to ions leaving the observation region before radiating. The upper limit is estimated to be 1.5 μsec for Nb II lifetimes with error less than 10% due to this effect. The levels studied in this initial experiment have lifetimes under 10 nsec; thus error due to ions leaving the observation region before radiating is negligible.

Figure 2(a) is a time-resolved laser-induced fluorescence decay curve generated by exciting the z^5G_5 level of Nb II with laser radiation at 313.079 nm. Figure 2(b) is a semilogarithmic plot of the decay curve. Radiative lifetimes of seven levels from the lowest two odd-parity terms of Nb II are reported in Table I. Each lifetime represents the average of ten or more decay curves. The standard deviation of the distribution is typically 5% or less. We quote overall uncertainties of $\pm 5\%$ for the Nb II lifetimes. Collisional quenching is not a problem in this experiment because the scattering chamber pressure is 10^{-4} Torr of Ar. Radiation trapping is not a problem in this experiment because the ion beam is optically thin. The most likely source of systematic error is due to the finite bandwidth of the detection apparatus.

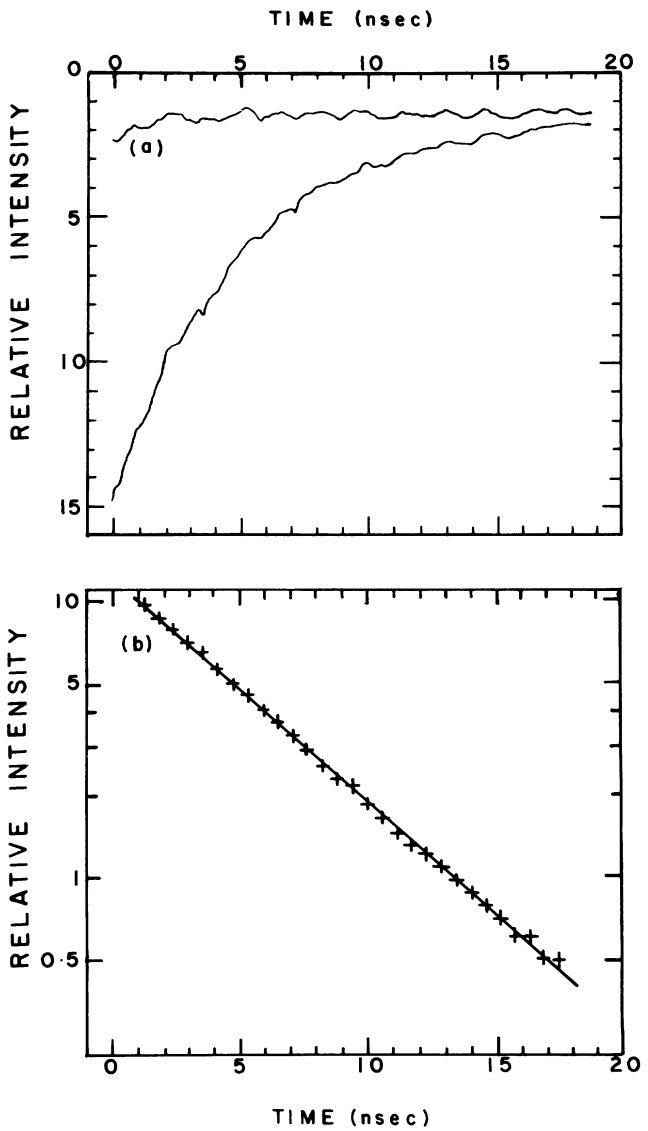

FIG. 2. (a) Time-resolved laser-induced fluorescence signal. The lower trace is the photomultiplier signal observed with the frequency doubled dye laser tuned to the Nb II transition at 313.079 nm. The upper trace is the signal observed with the dye laser tuned slightly off the transition. (b) Semilogarithmic plot of the difference between the upper and lower trace of (a). The solid line corresponds to a lifetime of 5.26 nsec. 0 nsec in the time scale of the figure corresponds to 5 nsec after the termination of the laser pulse.

The detection system is composed of a photomultiplier, a cable delay, and a boxcar averager. The bandwidth of the entire detection system is verified to be adequate for lifetimes as short as 7 nsec by studying the $4s\ ^2S_{1/2}$ level of Al I.[2] This level has a well-established lifetime of

TABLE I. Radiative lifetimes in Nb II.

		Energy (cm^{-1})	Excitation line (nm)	Lifetime (nsec) This expt.	Ref. 7
				(±5%)	(+100%-50%)
$4d^3(a^4F)5p$	z^5G_3	33919	319.498	6.2	12.5
	z^5G_4	34632	316.340	5.8	12.7
	z^5G_5	35474	313.079	5.3	10.8
	z^5G_6	36455	309.418	5.0	9.3
$4d^3(a^4F)5p$	z^3D_1	34886	309.919	5.5	5.0
	z^3D_2	35521	307.687	5.7	5.9
	z^3D_3	36553	302.844	5.0	5.4

6.78 ± 0.22 nsec.[8] We measure 6.71 ± 0.14 nsec. We have also studied the $z^4F_{9/2}$ level of Fe II. Hannaford and Lowe have recently determined the lifetime of this Fe II level to be 4.1 ± 0.3 nsec.[9] We measure 3.9 ± 0.2 nsec.

There are no other direct radiative lifetime measurements in Nb II. Our lifetimes are compared in Table I with lifetimes derived from the CB transition probabilities.[7] These investigators quote an uncertainty of ± 0.29 on $\log_{10}(gf)$ values determined from emission measurements on intense arcs. The CB lifetimes are determined by summing gA values for all the radiative decay channels tabulated for each level. The lifetimes determined by summing gA values are actually upper limits due to the possibility of strong infrared transitions not observed by Corliss and Bozman. It seems most likely that the decay of these levels is dominated by the strong UV transitions. Our lifetimes indicate that the CB gA values for transitions from the z^3D levels are correct and that the CB gA values for transitions from the z^5G levels are too small by a factor of 2. An improved transition probability for an emission line from one of the levels in Table I may be derived using the relation

$$gA = gA_{CB}(\tau_{CB}/\tau_{expt}) \ , \quad (1)$$

where gA is the improved transition probability, gA_{CB} is a transition probability from Corliss and Bozman's monograph, τ_{CB} is the lifetime derived Corliss and Bozman's data, and τ_{expt} is our experimental lifetime.

Five lines from the levels in Table I have been identified in the solar spectrum by Hauge and Youssef.[6] Unfortunately, there is some doubt about the accuracy of the equivalent widths of the Nb lines determined by these authors.[3] We suggest that a redetermination of the equivalent widths of the Nb II lines in the solar spectrum combined with the improved transition probabilities reported in this Brief Report will provide an important check on the solar abundance of Nb.

In summary, we describe a pulsed ion source suitable for laser spectroscopy on ions of any metallic element. We report the first direct radiative lifetime measurements in Nb II. The lifetimes are measured using time-resolved laser-induced fluorescence on an Nb II ion beam.

ACKNOWLEDGMENTS

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