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Radiative lifetimes in Zr I

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Radiative lifetimes of 11 levels from the $z\,^3G$, $y\,^5G$, and $y\,^3G$ terms of Zr I are reported. These lifetimes are measured using time-resolved laser-induced fluorescence on a Zr atomic beam.

Recently several new methods of measuring radiative lifetimes in refractory metal atoms have been reported[1,2] and applied to Zr I. The first of these methods involves bombarding a refractory metal surface in high vacuum with a pulsed heavy-ion beam.[1] Some of the sputtered metal atoms are electronically excited during the sputtering process. The intensity of radiation at one wavelength is measured as a function of time after termination of the ion beam pulse to determine a radiative lifetime. The second method involves producing free refractory metal atoms in a hollow cathode discharge.[2] Time-resolved laser-induced fluorescence is performed in the afterglow of the discharge to determine a radiative lifetime.

Table I summarizes the Zr I lifetime measurements reported in Refs. 1–3. Ramanujam[1] and Andersen, Ramanujam, and Bahr[3] reported Zr I lifetimes with uncertainties of ±4 to ±6%. Hannaford and Lowe[2] reported Zr I lifetimes with uncertainties of ±2 to ±4%. Unfortunately the measurements of Ramanujam and of Andersen, Ramanujam, and Bahr disagree with the measurements of Hannaford and Lowe by more than a factor of 3 in all cases where a comparison can be made.

Radiative lifetimes and line strengths of Zr I are of particular interest for several reasons. The measurements of Ramanujam and of Andersen, Ramanujam, and Bahr were used to check earlier Zr solar abundance determinations.[3] The measurements of Hannaford and Lowe were subsequently used in a precision Zr solar abundance determination.[4] Refractory metal atoms, sputtered from the walls of confinement devices, cause radiative cooling of thermonuclear plasmas. Atoms are sputtered from the walls predominantly as neutrals. Laser-induced fluoresence is used to study plasma-wall interactions in these machines.[5] Absolute line strengths are necessary if absolute metal-atom densities are to be obtained using laser-induced fluorescence.

Additional independent measurements of the Zr I lifetimes reported in Refs. 1–3 are desirable because of the substantial disagreements. We have performed such measurements using time-resolved

TABLE I. Radiative lifetimes of Zr I levels.

Level	Energy (cm^{-1})		Lifetime (ns)		This expt.
		Ref. 1	Ref. 3	Ref. 2	(±5%)
$4d^25s(a\,^4F)5p\ z\,^3G_3$	21 849	440 ±25		123±2	124
$z\,^3G_4$	22 144	380 ±15		111±2	118
$z\,^3G_5$	22 564	450 ±25		107±2	112
$4d^3(b\,^4F)5p\ y\,^5G_2$	25 630			12.3±0.3	12.0
$y\,^5G_3$	25 972			13.3±0.3	13.7
$y\,^5G_4$	26 343		41±2	13.1±0.3	13.6
$y\,^5G_5$	26 766		40±2	13.3±0.3	12.8
$y\,^5G_6$	27 215			12.0±0.5	11.1
$4d^3(b\,^4F)5p\ y\,^3G_3$	25 230			16.2±0.3	16.3
$y\,^3G_4$	26 012			21.5±0.4	21.5
$y\,^3G_5$	26 434			27.6±0.6	28.8

laser-induced fluorescence on a Zr atomic beam. The results are reported in Table I. The method we use has the advantages of both of the previously described methods and the disadvantages of neither. The pulsed ion beam technique of Ramanujam has the disadvantage that it uses a nonselective method of excitation. Although predominantly low-lying levels are excited, there is the possibility of problems due to radiative cascade from higher levels. There is, however, no evidence for cascade problems in the Zr measurement performed using this technique.[1,3] The pulsed ion beam technique has the advantage that the lifetime measurements are performed in high vacuum, therefore collisional quenching is not a problem. The technique of Hannaford and Lowe, involving time-resolved laser-induced fluorescence in a hollow cathode afterglow, has the advantage that it uses a highly selective means of excitation. A narrow-band pulsed dye laser excites a single fine-structure level, hence cascades from higher excited levels are definitely not a problem. The method of Hannaford and Lowe has the disadvantage that the lifetimes are not measured in a high vacuum. Care must be taken to correct for collisional quenching of the excited atoms.

The technique we use, time-resolved laser-induced fluorescence on an atomic beam, has the advantage that it uses a highly selective method of excitation. Cascades from higher excited levels are definitely not a problem. It also has the advantage that the excited atoms are in a nearly collision-free environment. No corrections for collisional quenching are necessary.

Figure 1 is a schematic of the experiment. The hollow-cathode effusive beam source is based on a low-pressure (≤ 0.25 Torr), large-bore (2.7 cm) hollow cathode developed for Doppler-free spectroscopy. This device has been described elsewhere.[6] The hollow cathode is modified for use as a beam source by closing one end of the cathode except for a 1-mm diameter nozzle, and by providing a continuous source of Ar, the sputtering gas. The hollow cathode is operated at a current of 20 to 200 mA. The uncollimated Zr beam emerges from the nozzle into a scattering chamber pumped by a 6 in. diffusion pump. The background Ar pressure in the scattering chamber when the hollow cathode is operating is approximately 10^{-4} Torr. The Zr beam is crossed by a pulsed dye laser beam 1.0 cm from the nozzle. The Zr beam, the dye laser beam, and the axis of the fluorescence detection system are mutually orthogonal. Several sets of light baffles are used to minimize scattered laser light. They are located before and after the Brewster windows that pass the dye laser beam into and out of the scattering chamber.

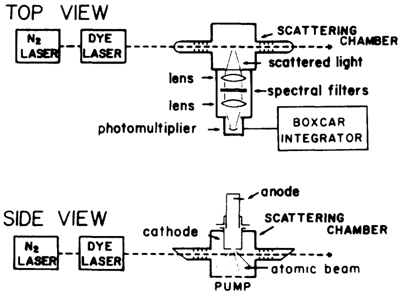

FIG. 1. Schematic of the experiment.

A pulsed dye laser, pumped by a N_2 laser, is used in this investigation. The N_2 laser produces a 0.5-MW pulse of 337.1-nm radiation of 5-ns (FWHM) duration.[7] The dye laser produces a pulse of tunable radiation (360 – 900 nm) of 3-ns (FWHM) duration.[8] The dye laser pulse begins and ends very abruptly as the laser crosses threshold. The dye laser provides impulse excitation to levels with lifetimes longer than a few nanoseconds. The dye laser bandwidth, 1 cm^{-1}, is sufficient to isolate a single fine-structure level. Thus, the fluorescence decay signal is not complicated by cascade from higher excited levels.

The fluorescence detection system consists of two $f/1$ lens with spectral filters between them, followed by a RCA 1P28A photomultiplier. In this investigation only low-lying odd-parity levels of Zr are studied. Thus, only a minimum of spectral filtering, provided by Corning dye filters, is necessary to block the cascade fluorescence involving lower levels. The bias resistors for the 1P28A photomultiplier are bypassed with capacitors to ensure good linearity at large peak currents. No loss of linearity for peak photomultiplier currents less than 20 mA is detectable. A single-photon spike, with a 2-ns (FWHM) duration, represents the impulse response function of the photomultiplier. A P.A.R. model 162 scanning boxcar integrator is used to provide temporal resolution of the fluorescence. The aperture duration of the boxcar integrator is adjustable; the minimum duration is 75 psec. Three pair of Helmholtz coils insure that the magnetic field in the scattering chamber is less than 0.05 G.

The Zr I levels studied in this investigation are excited from levels of the $4d^2 5s^2 a\ ^3F$ term and the

$4d^3(b\,^4F)5sa\,^5F$ term using dye laser radiation at the appropriate wavelengths between 360 and 500 nm. The $a\,^5F_5$ level is 5889 cm^{-1} above the ground level. The atomic beam from the hollow cathode is rich in metastable atoms.

Each radiative lifetime determined in this investigation is a mean of ten or more fluorescence decay curves. The standard deviation of a distribution of 10 or more measurements is typically ±5%. The standard deviation of a mean is less than ±2%. We have searched for systematic effects due to radiation trapping, collisional quenching, and stray magnetic fields in our radiative lifetime experiments.[9–11] No systematic effects have been detected. Radiation trapping is not observed on the $a\,^7S - z\,^7P$ resonance multiplet of Mo I which produces fluorescence signals sufficient to saturate the photomultiplier, and 10 to 100 times stronger than fluorescence signals in Zr.[9] The ground level of Mo has a proportionally larger population in our Mo atomic beam than the ground level of Zr in our Zr atomic beam because there are no metastable levels in Mo I below 10 000 cm^{-1}. Collisional quenching is not observed, even for levels with microsecond lifetimes.[10] The absence of collisional quenching is consistent with the 10^{-4}-Torr-Ar pressure in the scattering chamber. The agreement of zero-field and high-field measurements on levels with microsecond lifetimes confirms that the precession of polarized atoms in stray magnetic fields is not a problem.[11]

We routinely quote a total uncertainty of ±5% on our lifetime measurements.[9,10] We suggest, on the basis of the agreement between our Zr I measurements and those of Hannaford and Lowe, that the ±5% figure is conservative. The average difference between our lifetimes and those of Hannaford and Lowe is +0.9%, the root-mean-squared difference is 4%.

In summary, we have measured the radiative lifetimes of 11 levels from the $z\,^3G$, $y\,^5G$, and $y\,^3G$ terms of Zr I using time-resolved laser-induced fluorescence on an atomic beam. Our measurements are in excellent agreement with those of Hannaford and Lowe. Hannaford and Lowe's measurements and our measurements are in substantial disagreement with earlier lifetime measurements in Zr I performed using a pulsed ion beam technique.

Note added. We are informed of additional measurements in Zr I which were in press when our manuscript was submitted. The additional measurements also support Hannaford and Lowe. Poulsen, Andersen, Bentzen, and Nielsen report a lifetime for the $z\,^3G_5$ level of 118 ±5 ns.[12] Poulsen, Andersen, Bentzen, and Koleva report lifetimes for the $y\,^5G_3$, $y\,^5G_4$, and $y\,^5G_5$ levels of 12, 13, and 13 ns (±15%), respectively.[13]

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