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Radiative lifetime measurements of odd-parity highly-excited levels of Sn I by time-resolved laser spectroscopy*

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Natural radiative lifetimes of five higher-lying odd-parity levels 5p7s $^3P^o_1$, 5p5d $^1P^o_1$, 5p6d $^3F^o_2$, $^3D^o_1$ and $^3F^o_3$ in neutral tin are measured by the time-resolved laser-induced fluorescence (TR-LIF) technique and the atomic beam method. All these lifetimes are not longer than 100 ns and they are found to be shorter than the lifetimes of even-parity levels in the same energy region. The results reported in this paper provide important transition parameters for highly-excited atomic Sn, which may be useful for theoretically calculating excited heavy atoms.

Keywords: radiative lifetime, atomic Sn, highly-excited level, laser spectroscopy
PACC: 3270F

1. Introduction

Neutral tin (Sn I, $Z = 50$), which is the second heaviest carbon-group element, has a $5s^25p^2$ ground configuration, and its singly excited configurations consist of an excited electron outside of a 5p-electron ionic core with two ionization limits: $^2P^o_{1/2}$ (59232.69 cm^{-1}) and $^2P^o_{3/2}$ (63484.18 cm^{-1}).[1] Due to its rather simple structure, the energy levels of Sn I have been investigated adequately. Moore summarized the early data,[2] later on, Brill[3] and Wilson[4] studied the atomic structure of Sn I by using an arc discharge and absorption technique, respectively, but their results were not published. Brown *et al* [1] reported on the high-resolution absorption spectrum of Sn I in the region between 158 and 204 nm and determined many odd-parity levels. Recently, Nadeem *et al* [5–7] investigated numerous even-parity levels with $J = 1$, 2 from 5pnp and 5pnf configurations as well as many autoionizing levels by using a two-step excitation technique. It should be mentioned that Jin *et al* [8] also measured some 5pnp and 5pnf even-parity Rydberg levels with $J = 0$–3 by utilizing the resonant multi-photon ionization method and time-of-flight mass spectroscopy, but the results were not published. Dembczynski and Rebel[9] and Dembczynski and Wilson[10] theoretically investigated the odd-parity levels and the $5s5p^3$ levels of Sn I in detail.

Although the energy levels have been extensively investigated, the experimental determination of Sn I lifetimes is limited to low-lying odd-parity levels and a few highly-excited even-parity levels, and hence the lifetime data of this atom are still extremely scarce. For odd-parity levels, in the previous work, the lifetimes of the 5p6s $^1P^o_1$ and $^3P^o_{1,2}$, 5p5d $^3D^o_{1,2,3}$ levels have been measured by Penkin and Slavenas[11] by using the hook method. Lawrence[12] investigated the lifetimes of the 5p6s $^3P^o_{1,2}$ levels by the phase-shift technique. By the zero-field level-crossing (Hanle effect) method, Brieger and Zimmermann,[13] and Zafra and Marshall[14] measured the lifetimes of the 5p6s $^3P^o_1$ level, respectively, while Holmgren and Svanberg[15] investigated the lifetimes of the 5p6s $^1P^o_1$, $^3P^o_{1,2}$, and 5p5d $^3D^o_{1,2,3}$ levels. In 1985, Gorshkov and Varolainen[16] measured the lifetimes of the 5p6s $^1P^o_1$, $^3P^o_{0,1,2}$, and 5p5d $^3D^o_{1,2,3}$ and $^3F^o_{2,3}$ levels by using the multichannel method of retarded coincidences. It is remarkable that among the 9 levels measured by these different techniques, only the 5p5d $^3F^o_2$ lifetime reaches 24 ns while the others are shorter than 7 ns.

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For even-parity levels, lifetimes of 9 levels of 5p7p configuration have been reported by us recently.[17] To the best of our knowledge, no other work has been published in the literature.

The reasons why the experimental lifetime data are so rare are as follows: (1) a high-temperature source is necessary for obtaining an atomic beam with sufficient vapour density; (2) for odd-parity levels, almost all the excitations from the ground level need a vacuum ultraviolet laser which is rather difficult to obtain while, for even-parity levels, the intermediate resonance levels used for a two-step excitation can be excited only by UV laser light and their lifetimes are only a few nanoseconds.

Although the lifetimes of Sn I levels are difficult to measure, they are very useful in some fields. (1) In atomic physics, where in order to further gain an insight into the properties of excited states of Sn I, additional atomic structure data, such as natural radiative lifetimes, Landé g_J factors as well as hyperfine structure information, are required. (2) In astrophysics, where Sn isotopes are produced by the p, s and r processes, and Sn has been identified from the photospheric solar spectrum and the spectra of some chemically peculiar hot stars.[18] It is well-known that the determination of solar and stellar abundances for many chemical elements is limited by the accuracy of the available atomic transition probabilities[19] which can be deduced from accurate radiative lifetimes and accurate branching fractions obtained experimentally or theoretically. So a necessary starting step for a quantitative investigation of stellar spectra consists in determining radiative lifetimes accurately. (3) In environmental protection. Recently, with the development of lead-free techniques, the utilization of tin, which has been regarded as an innocuous substitute, has become more extensive.[20] In the present paper, we report on the lifetime measurements of five higher-lying odd-parity levels 5p7s $^3P_1^o$, 5p5d $^1P_1^o$, 5p6d $^3F_2^o$, $^3D_1^o$ and $^3F_3^o$ in Sn I by using the TR-LIF technique in an atomic beam.

2. Experimental setup

The experimental setup used for lifetime measurement is shown in Fig.1. A high-temperature oven was mounted at the bottom of a vacuum chamber for providing an atomic beam with a sufficient vapour density. Its operating temperature can reach up to 1700 K. The oven was surrounded by heating molybdenum wires. The oven system is made of corundum. There is a 1.5 mm-diameter hole on the cover of the crucible for generating an atomic beam with a low collimation ratio.

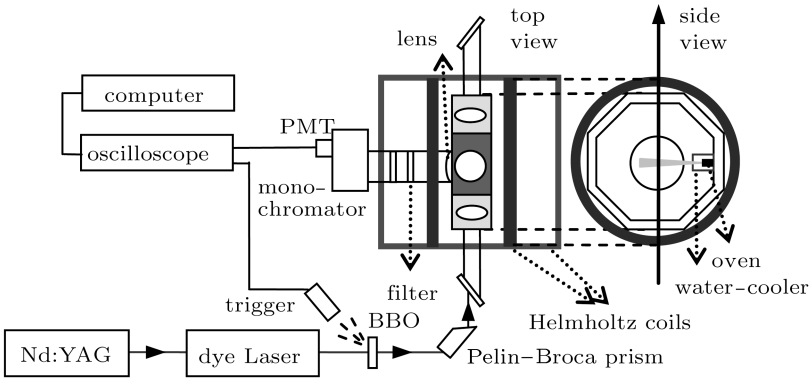


Fig.1. Experimental setup used for lifetime measurement.

A linearly polarized dye laser (Sirah Cobra-Stretch) pumped by a Q-switched Nd:YAG laser (Spectra-Physics Quanta-Ray Pro-Series) working at a 10 Hz repetition rate and with about 8 ns pulse duration was used for excitation. The linewidth of the dye laser was about 0.08 cm^{-1}. The light from the dye laser was focused on a BBO type-I crystal to generate a frequency-doubled light which induced the transi-

tions from the ground and metastable states to the selected odd-parity levels. The laser beam encountered the vertical atomic beam at the centre of the vacuum chamber. Following the excitation, the fluorescence signal was focused onto a grating monochromator by a fused silica lens and then detected by a photomultiplier tube (PMT) (Hamamatsu R3896) in a direction perpendicular to the laser and the atomic beam. A 500 MHz digital oscilloscope (Tektronix TDS 620B) was used to record the time-resolved photocurrent signal from the PMT. The oscilloscope was connected through a GPIB cable to a computer where the signal could be analysed.

A pair of Helmholtz coils was used to generate a strong magnetic field which could be up to 200 G in the direction of the fluorescence detection, thus it could wash out the quantum beats produced by the earth magnetic field in order to eliminate possible distortion of the decay curves. In the same way, another pair of coils was employed to neutralize the vertical component of the earth field. The two sets of coils were powered by two high-stability constant-current power supplies.

3. Results and discussion

In the present experiment, the Exalite 411 dye laser was used for exciting the high-lying odd-parity levels. The second harmonic of the dye laser was generated through a BBO crystal producing an ultraviolet laser in a tunable range from 206 to 210 nm. The 5p7s $^3P_1^o$ level was excited from the ground level, the 5p5d $^1P_1^o$ level from the 1691.8 cm^{-1} metastable level, and the 5p6d $^3F_2^o$, $^3D_1^o$ and $^3F_3^o$ levels from the 3427.7 cm^{-1} metastable level. The diagram of energy levels and the detailed excitation schemes related to the experiment are illustrated in Fig.2. In the measurement, some effects which can introduce additional errors into the results have to be carefully taken into account. The possible flight-out-of-view effect was checked by changing the slit width and the slit position of the monochromator along the propagating direction of the atomic beam. The effect of collision quenching was checked by changing the air pressure in the vacuum chamber and the oven temperature. In addition, the variation of atom number density by changing the oven temperature was used to check the effect of radiation trapping. As a result, it is found that the above effects are negligible since all the lifetimes measured in this experiment are not too long.

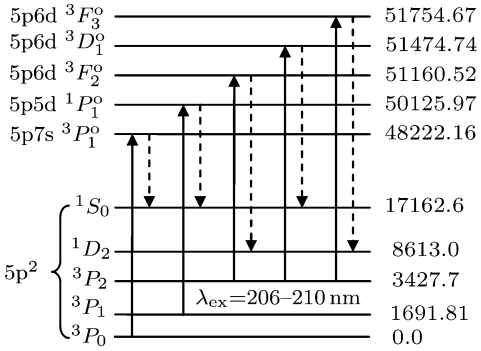


Fig.2. Partial energy-level diagram of the Sn atom and excitation schemes. The solid lines indicate the excitation pathways, and the dash lines show the fluorescence channels. Energy levels is in units of cm^{-1}.

In order to acquire a good signal, more than 2000 shots were averaged for each fluorescence decay curve. Lifetimes were evaluated by a least-squares exponential fitting to the decay part of the fluorescence curve. The starting point of this fitted decay part should be chosen so as to exclude the influence of the laser pulse. At least 10 curves were recorded for each level under different experimental conditions and the mean value was taken as the final lifetime result. A decay curve of the 5p7s $^3P_1^o$ level is shown in Fig.3 as an illustrative curve of this kind.

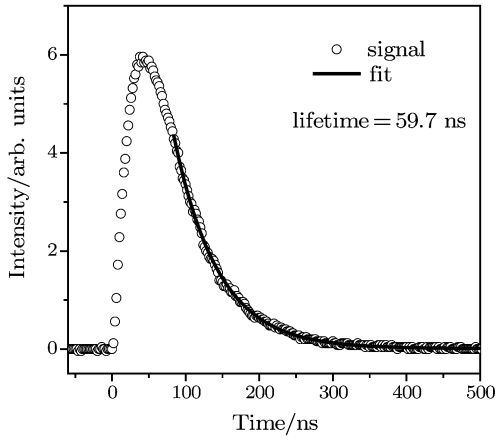


Fig.3. Typical fluorescence decay curve of the 5p7s $^3P_1^o$ level with an exponential fitting for lifetime evaluation.

For the 5p5d $^1P_1^o$ level, its lifetime is so short that it is comparable with the pulse duration of the exciting laser. For this case, the lifetime could be evaluated not by a direct exponential fitting but by a deconvolution procedure. In fact, at time t, the real fluorescence intensity $I(t)$ acquired by the detection system is related

to the laser pulse intensity $L(t)$ and the instrumental response function $R(t)$ through the expression[21]

$$I(t) = \int_{-\infty}^{t} L(t')R(t')f(t-t')\mathrm{d}t', \qquad (1)$$

where $f(t)$ is the fluorescence decay function reflecting decay behaviour of the upper level population, which generally is a pure exponential. $L(t')R(t')$ corresponds to the laser pulse function recorded by the detection system, so the true fluorescence decay function $f(t)$ can be evaluated by deconvolving the recorded fluorescence curve $I(t)$. When the level lifetime is much longer than the laser pulse duration, the major decay part of the fluorescence curve $I(t)$ will not be affected by the term $L(t')R(t')$. The recorded fluorescence curve for the 5p5d $^1P_1^o$ level together with a deconvolution fitting curve is shown in Fig.4. The lifetime of this level is determined to be 8.8 ns from the deconvolution calculations.

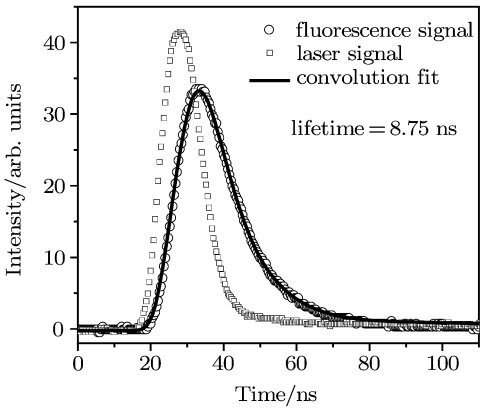


Fig.4. Fluorescence decay curve of the 5p5d $^1P_1^o$ level together with the fitted convolution curve between the laser pulse and an exponential with a decay constant of 8.8 ns.

The measured lifetimes for the odd-parity 5p7s $^3P_1^o$, 5p5d $^1P_1^o$, 5p6d $^3F_2^o$, $^3D_1^o$ and $^3F_3^o$ levels are listed in Table 1. The quoted error bars reflect both the statistical uncertainties from different recordings and the systematic errors resulting from the fitting procedure. These five lifetime values do not exceed 100 ns and the shortest one is only around 9 ns. In this energy region for measurement (48000 to 52000 cm^{-1}), the lifetimes of all four even-parity levels have been previously measured by us and they were located between 145 and 262 ns.[17] Obviously, the odd-parity levels in this region will decay faster than the even-parity levels. This is easy to understand when considering that the levels from the ground configuration are of even parity and will provide short-wavelength electric-dipole-allowed decay channels.

Table 1. Measured lifetimes of the odd-parity 5p7s $^3P_1^o$, 5p5d $^1P_1^o$, 5p6d $^3F_2^o$, $^3D_1^o$ and $^3F_3^o$ levels of neutral tin.

E/cm^{-1}[1]	term[1,9]	excitation		lifetimes/ns
		origin	λ/nm	
48222.16	$5p_{1/2}7s\ ^3P_1^o$	$5p^2\ ^3P_0$	207.374	61(2)
50125.97	$5p_{1/2}5d\ ^1P_1^o$	$5p^2\ ^3P_1$	206.466	8.8(6)
51160.52	$5p_{1/2}6d\ ^3F_2^o$	$5p^2\ ^3P_2$	209.499	89(5)
51474.74	$5p_{1/2}6d\ ^3D_1^o$	$5p^2\ ^3P_2$	208.129	49(8)
51754.67	$5p_{3/2}6d\ ^3F_3^o$	$5p^2\ ^3P_2$	206.924	61(4)

The highest level measured in the present experiment is 51754.67 cm^{-1}. The higher levels above it cannot be excited by the one-step excitation technique from the ground level or the lower metastable levels without using a vacuum ultraviolet laser. Between the investigated levels in this paper, the level 49487.13 cm^{-1} can also be excited by the Exalite 411 dye laser, but its fluorescence signal is too weak to be used to evaluate a lifetime result.

Figure 5 presents all the measured lifetimes of 14 odd-parity levels in Sn I, for which the lower nine levels are cited from Refs.[15] and [16] and the other higher 5 levels are obtained from the present study. It should be pointed out that in Refs.[2] and [9], the assignments of spectral terms for the levels 47487.70 and 51754.67 cm^{-1} are interchanged while for the other 12 levels they are consistent with each other. Here the assignments to the spectral terms of the former two

levels are adopted from Ref.[9] because more complex configuration interactions on higher-lying levels make, to a certain extent, the transition rules ineffective and hence for the determination of total orbital angular momentum L in spectral terms the theoretical analysis results of atomic structure must be used as a reference. It can be seen from Fig.5 that the lifetimes of the 5p7s and 5p6d levels are of the same order of magnitude, just as in the case of the 5p6s and 5p5d levels. A comparison between the 5p5d level and the 5p6d level indicates that the $^3F_2^o$ level has a longer lifetime.

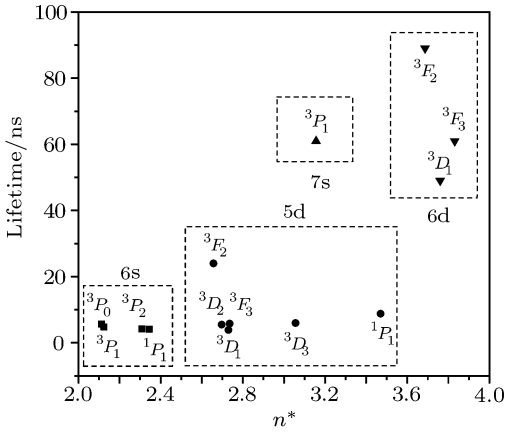


Fig.5. Experimental lifetimes versus the effective quantum number of the corresponding levels. The lifetime data of the lowest nine levels are cited from Refs.[15] and [16].

4. Conclusions

The experimental lifetimes of the five high-lying odd-parity levels 5p7s $^3P_1^o$, 5p5d $^1P_1^o$, 5p6d $^3F_2^o$, $^3D_1^o$ and $^3F_3^o$ in Sn I are determined by the time-resolved laser-induced fluorescence technique and the atomic beam method. All these lifetimes are not longer than 100 ns. Based on the known lifetime data of odd-parity levels, it could be concluded that the lifetimes of the levels from the 5p7s and the 5p6d configurations will be of the same order of magnitude, which is similar to the case for the 5p6s and the 5p5d configurations.

References

[1] Brown C M, Tilford S G and Ginter M L 1977 *J. Opt. Soc. Am.* **67** 607

[2] Moore C E 1958 *Atomic Energy Levels* Vol. III Circ. of Natl. Bur. Std. (US), No. **467** (Washington, D. C.: U.S. Government Printing Office) p74

[3] Brill W G 1964 *The Arc Spectrum of Tin* Ph. D. Thesis (Purdue University)

[4] Wilson J W 1964 *The Atomic Absorption Spectra of Silicon, Germanium, Tin and Lead* Ph. D. Thesis (Imperial Coll. of Sci. and Tech. of London)

[5] Nadeem A, Ahad A, Bhatti S A, Ahmad N, Ali R and Baig M A 1999 *J. Phys.* B **32** 5669

[6] Nadeem A, Bhatti S A, Ahmad N and Baig M A 2001 *J. Phys.* B **34** 2407

[7] Nadeem A, Bhatti S A, Ahmad N and Baig M A 2000 *J. Phys.* B **33** 3729

[8] Jin M X, Ding D J, Liu H and Pan S F 1990 *Inst. Phys. Conf. Ser.* **114** 235

[9] Dembczynski J and Rebel H 1984 *Physica* B&C **125** 341

[10] Dembczynski J and Wilson M 1988 *Z. Phys.* D **8** 329

[11] Penkin N P and Slavenas I Yu-Yu 1963 *Opt. Spectrosc.* **15** 83

[12] Lawrence G M 1967 *Astrophys. J.* **148** 216

[13] Brieger M and Zimmermann P 1967 *Z. Naturforschg.* A **22** 2001

[14] Zafra R L and Marshall A 1968 *Phys. Rev.* **170** 28

[15] Holmgren L and Svanberg S 1972 *Phys. Scr.* **5** 135

[16] Gorshkov V N and Verolainen Ya F 1985 *Opt. Spectrosc.* **59** 1157

[17] Zhang Y, Xu J X, Zhang W, You S, Ma Z G, Han L L, Li P F, Sun G J, Jiang Z K, Enzonga Yoca S, Quinet P, Biémont É and Dai Z W 2008 *Phys. Rev.* A **78** 022505

[18] Adelman S, Bidelman W P and Pyper D 1979 *Astrophys. J. Suppl.* **40** 371

[19] Asplund M, Grevesse N and Sauval A J in: 2005 Bash F N and Barnes T G (editors) *ASP Conference Series* (San Francisco: ASP) **336** 25

[20] Pennington J N 1999 *Mod. Met.* **55** 38

[21] Zuker M, Szabo A G, Bramall L, Krajcarski D T and Selinger B 1985 *Rev. Sci. Instr.* **56** 14