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Radiative lifetimes of some excited states of neutral xenon

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Abstract. The radiative lifetimes of ten levels belonging to the nf ($n = 4-7$) and three levels belonging to the ns ($n = 8$, 9) configuration of XeI have been measured using the high frequency deflection technique together with a delayed coincidence single-photon counting arrangement. Lifetimes of some of the levels have been measured for the first time. The results have been compared with other experimental and theoretical values.

PACS. 32.70.Cs Oscillator strengths, lifetimes, transition moments

1 Introduction

The knowledge of the radiative lifetime of excited atomic levels is useful for a better understanding of the atomic structure. The lifetime together with branching ratios provides information about transition probability and hence oscillator strength having important applications in different branches of physics [1]. Due to the use of xenon in the discharge medium (high efficiency light sources, plasma displays) or in the outer layer of fusion reactors, accurate experimental data are necessary in order to decide which theoretical models and methods are best suited for calculating the radiative constants of xenon atoms where different theoretical approaches led to different results. It is also important and worthwhile to carry out experimental measurement of atomic lifetimes for the levels where no theoretical data exist. Many experiments were performed in order to determine the radiative lifetimes of xenon (XeI) excited states. Measurements were carried out by Verolainen and Osherovich [2], Jimenez and Campos [3] and Martin et al. [4] using the delayed coincidence technique. Horiguchi et al. [5], Inoue et al. [6] and Whitehead et al. [7] also investigated some of the levels using the laser induced fluorescence technique. In addition, there are reports of measurements by Kazantsev et al. [8] and Gorny et al. [9] using the Hanle effect. However, most of the measurements concentrate on the $6p$ and $7p$ states since the strong transitions in the XeI spectrum belong to these configurations. There is a shortage of experimental information regarding the lifetimes of $5p^5nf$ ($n = 4-7$) and $5p^5ns$ ($n = 8$, 9) levels of XeI. The only previous measurement for some of the nf levels of XeI was done by Verdugo et al. [10] using the electron excitation delayed coincidence technique. There are no experimental data for the $8s$ levels available and for the $9s[3/2]_1$ level only a single measurement was performed by Jimenez and Campos [3]. Theoretical data for these levels are also meager. There are calculations carried out by Loginov and Gruzdev [11] for the $4f$, $8s$, and $9s$ levels. Verdugo et al. [10] and Jimenez and Campos [3] also calculated lifetimes of some of the nf ($n = 4-7$) and ns ($n = 8$, 9) levels, respectively. There are no theoretical data for some of the $6f$ and $7f$ levels. Therefore, we undertook an experimental investigation for ten levels belonging to the nf ($n = 4-7$) and for three levels belonging to the ns ($n = 8$, 9) configurations of neutral xenon. For the $6f[7/2]_3$, $6f[9/2]_5$, $7f[7/2]_3$, $8s[3/2]_1$ and $8s[3/2]_2$ levels no experimental results have been published to our knowledge until now. Lifetimes were measured through transitions having wavelengths ranging from 5162 to 8058 Å.

2 Experimental method

In the present work we used the high-frequency deflection (HFD) technique [12] in which excitation of the levels was produced by high-energy pulsed electron impact. The corresponding decay had been followed by a delayed-coincidence method using the single-photon counting arrangement. The experimental set-up was similar to that used in a previous investigation [13] with the following improvements in the experimental device. A condensing lens (Oriel 77260 lens-filter-shutter assembly) has been used for focusing the emitted radiation exactly onto the monochromator entrance slit. It causes an enhancement in the intensity of the emitted spectral line and hence increases the spectral resolution, because measurements can be done with a narrow slit width of spectrometer.

Thus the problem of line blending is reduced. Measurements can also be done at a low gas pressure, reducing in

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Table 1. Lifetimes (ns) of some $5p^5nf$ ($n = 4$–7) and $5p^5ns$ ($n = 8$–9) levels of XeI.

Level	Wavelength (Å)	Experimental values: Present work	Experimental values: Verdugo et al. Ref. [10]	Experimental values: Jimenez & Campos Ref. [3]	Theoretical values: Verdugo et al. Ref. [10]	Theoretical values: Loginov & Gruzdev Ref. [11] τ_{rv}	Theoretical values: Loginov & Gruzdev Ref. [11] τ_{ma}	Theoretical values: Jimenez & Campos Ref. [3]
$4f[3/2]_1$	6827.3	55.4 ± 3.8	63 ± 7		50.6	14.8	30.9	
$5f[5/2]_3$	7643.9	89.7 ± 6.2	104 ± 6		97.1			
$5f[9/2]_4$	8057.3	98.1 ± 6.7	101 ± 7		91.8			
$6f[3/2]_1$	5392.8	128.3 ± 8.9	148 ± 12		160.9			
$6f[5/2]_2$	5688.4	137.8 ± 9.6	153 ± 12		169.7			
$6f[7/2]_3$	7783.7	147.5 ±10.3						
$6f[9/2]_5$	6872.1	120.2 ± 8.4						
$7f[3/2]_1$	5162.7	211.3 ±14.7	245 ± 23		249.9			
$7f[5/2]_2$	6412.4	198.6 ±13.8	242 ± 15					
$7f[7/2]_3$	7319.9	177.2 ±12.4						
$8s[3/2]_1$	7802.7	71.1 ± 4.9				19.4	66.0	97.5
$8s[3/2]_2$	7386.0	78.9 ± 5.5				92.3	108	107.8
$9s[3/2]_1$	6533.2	142.7 ± 9.9		180.0		36.0	33.4	180.0

τ_{rv} – calculated from geometric mean of the line strength in the length and velocity form in the single configuration approximation.
τ_{ma} – calculated from the multiple configuration approximation.

this way the pressure-dependent effect. Some modification in the electronic circuit of the electron beam sweeping system has been done to achieve an extended range of sweep frequency from 250 kHz to 2 MHz. Thus, the repetition period can be varied from 500 ns to 4000 ns so that the decay curve can be followed for many lifetimes for investigation of long-lived cascade components. A focused 2 mA electron beam of an energy of 4 keV was produced by an electron gun. To obtain a pulsed beam of very short duration (2.2 ns) the dc beam was deflected by applying a fast rising voltage pulse across a narrow slit at a high frequency (1 MHZ). The pulsed electron beam interacted with the xenon molecules and was finally collected by a metal plate. The spectral lines of interest were selected by means of a 0.5 meter grating monochromator (Minuteman, model 305) with a resolution of 0.7 Å. Single photons were detected with a Hamamatsu R943-02 photomultiplier tube (PMT) cooled down to −20 °C to reduce the dark current. The tables of Striganov and Sventitskii [14] and Saloman [15] have been used to identify the measured transitions. The coincidence resolving time (FWHM) of the whole system was measured by the method described by Erman [12] and was found to be 3.2 ns. The time calibration of the system was accomplished using a time calibrator (Ortec 462). The decay curves were observed in the time range of 1000 ns and the time delay in the coincidence set-up was 1.6 ns per channel. The measurements were carried out at a few gas pressures ranging from 0.2 to 4 mtorr and a continuous gas flow was maintained during each measurement. No significant dependence of any of the measured lifetimes on pressure was observed within the experimental error. The lifetime of each level was determined from the corresponding decay curve by making a least squares fit to the experimental data convoluted with the known instrumental response function [16].

3 Results and discussion

The results of the present lifetime measurement of some XeI levels are presented in Table 1. It also includes the experimental and theoretical values obtained by other investigators. The lifetime values are the weighted average of three independent measurements for each level. The quoted errors include the statistical and systematic errors. In the present measurement, the statistical error is about 3%. The main sources of the systematic error are due to the following factors.

The variation of the width of the instrument response function: it may occur as a result of the variation of the width of the pulsed electron beam mainly due to a change in shape and size of the electron beam profile. Using highly stable voltage and current supplies for the beam focusing and sweeping system, the error has been restricted within 0.2%.

Uncertainty due to non-linearity of the measuring system and uncertainty in the time calibration: there may be some variations in the discriminator threshold of the constant fraction discriminator and the time to amplitude converter and in the gain of the fast amplifier used in the experiment. With modern electronic equipments (Ortec modules) the uncertainty introduced by the integral and differential non-linearity of the system including the pulse height analyzer is less than 0.2%, whereas the uncertainty in the time calibration is about 0.1%.

An error of 3% has been estimated (see below) due to the cascade feedings from higher levels. Lifetimes of different XeI levels were measured at gas pressures ranging from 0.2 to 4 mTorr. The pressure-induced change of lifetime is small for a measurement within this pressure range and the maximum error amounts to 0.5%.

Thus, the total error estimation includes the following uncertainties: counting statistics (3%), instrumental time resolution (0.2%), time calibration (0.1%), non-linearity of the electronic system (0.2%), cascading (3%) and pressure effects (0.5%).

To check the reliability of our measuring system we have measured first the lifetimes of $7p[1/2]_0$ and $7p[3/2]_2$ levels. These were measured earlier by Whitehead et al. [7] to a high degree of accuracy and the values reported there are 63.6 ± 4.9 ns and 108.0 ± 2.1 ns, respectively. Our measured lifetime values for the above two levels are 67.8 ± 5.1 ns and 114.1 ± 8.5 ns, respectively which are in good agreement with those of Whitehead et al. [7].

Below 30 000 Å, the lists of classified lines of Striganov and Sventitskii [14] and Saloman [15] for XeI were found to contain no line that appears due to transition from a higher level to any of the levels under investigation. An excitation spectrum (intensity vs. wavelength scan) has been recorded between 2000 to 9000 Å by using the monochromator at a resolution of 0.7 Å, in order to investigate whether the spectrum contains any lines other than those present in their lists. All the lines recorded in this spectrum were found in the lists of classified lines of Striganov and Sventitskii [14] and Saloman [15] for XeI. However, their lists contain many more lines which were not observed in the present experiment. It was not possible to examine the other cascading levels that decay by emitting photons having wavelengths below 2000 Å and higher than 9000 Å in this way because of the limited spectral range of the PMT. In the list of Saloman [15], beyond 30 000 Å, there are a few lines that might have cascading effects in our measurement. But it is seen from the work of Massey [17] that for excitation with electrons having energies well above the threshold, the higher levels are expected to be excited with relatively less probability as compared to the lower ones. As the excitation energy is 4 keV in the present measurement, the probability for cascade interference in lifetime determination is less severe as compared to the conventionally used electron beam in the low energy region. Thus, in the present case cascade feedings from higher levels do not seem to contribute to any significant error in the lifetime measurement. In our earlier measurement [13] for the lifetime of some $4p$ levels of ArII, some cascading lines were observed within 2000 to 9000 Å. But the maximum error in the determination of the lifetime due to cascade feedings was found to be 3%. The computer program [16] we used to determine the lifetime value for a level from its corresponding decay curve also analyzes the multi-exponential components. In the present work each of the decay curves was best fitted to a single exponential. However, since our detector is not sensitive in the region above 9000 Å and below 2000 Å, an error of 3% has been included in our estimation of the total systematic error. A typical decay curve for the $5f[9/2]_4$ level, measured at 8057.3 Å, is shown in Figure 1.

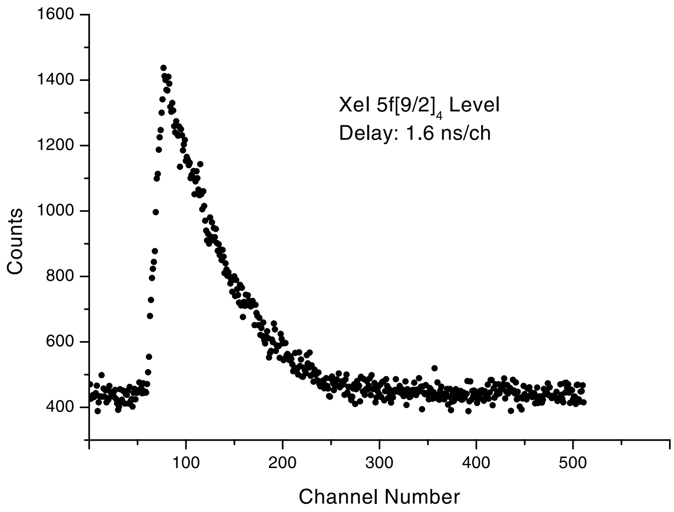

Fig. 1. The decay curve of the $5f[9/2]_4$ level of XeI measured at 8057.3 Å (gas pressure 6×10^{-4} Torr).

Except for the $4f[3/2]_1$, $5f[5/2]_3$ and $5f[9/2]_4$ levels the experimentally obtained values of Verdugo et al. [10] are larger than the corresponding values obtained in the present work. For $9s[3/2]_1$ level, the experimental lifetime measured by Jimenez and Campos [3] is also larger than ours. In the experimental set-up of Verdugo et al. [10] and Jimenez and Campos [3] the spectral resolutions were 6 Å and 3 Å, respectively, quite high in comparison to ours and for some levels line blending might have occurred affecting the accuracy in the lifetime measurement. In most cases, Verdugo et al. [10] could not resolve the nf ($n = 4{-}7$) fine structure components arising from levels of the same configuration with different values of the J quantum number. Furthermore, there are some neighboring XeI and XeII (singly ionized xenon) spectral lines very close to the primary lines (lines selected for lifetime measurement of the levels) and those lines might be responsible for the blending problem in their measurement. These lines (Å) are 4384.9, 4208.5, 4209.5 (XeII) for the $4f[3/2]_1$ level, 7642.0 (XeI) for the $5f[5/2]_3$ level, 7589.6 (XeI) for the $5f[9/2]_4$ level, 6922.2 (XeI) for the $6f[5/2]_2$ level, 5164.4, 5167.3 (XeI) and 5438.9, 5445.5 (XeII) for the $7f[3/2]_1$ level, and 6556.7, 6563.2 (XeII) for the $7f[5/2]_2$ level. Jimenez and Campos [3] determined the lifetime of $9s[3/2]_1$ level from the double exponential component of the decay curve for the $8p[3/2]_1$ level which was measured through the line having a wavelength 4116 Å. There were two XeI lines at 4113.5 and 4113.3 Å and those lines could not be resolved from the primary line at 4116 Å.

There are different theoretical approaches by several authors. The calculations of Verdugo et al. [10] and

Jimenez and Campos [3] were performed in the Coulomb approximation and *jk* coupling. However, for most of the levels their calculated values differ from our experimental values. This semi-empirical theory may not be appropriate to calculate lifetimes in xenon atoms because the possibility of configuration interaction (electron correlation) was ignored in their calculations, although there may be appreciable interaction between some XeI excited configurations. The calculations of Loginov and Gruzdev [11] were based upon an intermediate coupling scheme, using Hartree-Fock radial functions to compute the energy matrices for the electron-electron Coulomb interaction and the spin orbit interaction. Using only one configuration, they calculated the lifetimes in length-form τ_r, velocity-form τ_v and the geometric mean of both the values τ_{rv} (shown in Tab. 1). Furthermore, configuration mixing, present in the xenon atom, was taken into account by calculation with a set of eleven configurations which yielded τ_{ma} (shown in Tab. 1) in the geometric mean. But the calculated values differ widely from the lifetime values obtained in the present measurement. The large discrepancies between the different forms of the dipole matrix element occurring in their calculation may be due to a deficiency of the method used for taking account of the overlap of configurations and it should be removed in order to achieve a higher level of accuracy. The inclusion of a restricted set of eleven configurations in the calculation is possibly another cause for the deviation, because drastic changes in the calculated data often occur the more correlation configurations are included [18]. Thus new theoretical efforts are needed taking into account more precisely the effect of electron correlations.

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