 ©2009 Kvantovaya Elektronika and Turpion Ltd

PACS numbers: 32.50.+d; 41.75.Fr
DOI: 10.1070/QE2009v039n02ABEH013910

Population and relaxation kinetics of the $5d[3/2]_1$ level upon pulsed electron-beam excitation of pure xenon

I.A. Denezhkin, P.P. D'yachenko

***Abstract*. The time dependences of the spontaneous emission intensity at the $5d[3/2]_1 \rightarrow 6p[3/2]_1$ and $5d[3/2]_1 \rightarrow 6p[5/2]_2$ transitions at 2.03 and 1.73 μm, respectively, are studied in the pressure range from 0.1 to 100 Torr upon excitation by a nanosecond electron beam. It is shown that the population and relaxation of the $5d[3/2]_1$ level under these conditions is determined by its excitation and quenching by electrons of a recombining plasma. No plasma-chemical processes populating this level, in particular, a channel of formation of a molecular Xe_2^+ ion was found. The lifetime of the $5d[3/2]_1$ state of Xe atoms in the pressure range from 0.5 to 5 Torr is about 1 ns.**

Keywords: *xenon, electron beam, level lifetime.*

1. Introduction

Lasers on the $5d \rightarrow 6p$ transitions of atomic xenon, in which a He – Ar – Xe mixture can be pumped by an electric discharge, an electron beam or nuclear fission fragments are widely used to generate radiation in the near-IR region from 1.73 to 3.65 μm. It is known [1] that the He – Ar – Xe mixture is considered as a promising active medium for the development of high-power reactor-laser systems, in particular, pulsed systems (nuclear-pumped laser amplifiers) [2]. Of most interest are the $5d[3/2]_1 \rightarrow 6p[5/2]_2$, $5d[3/2]_1 \rightarrow 6p[3/2]_1$ and $5d[3/2]_1 \rightarrow 6p[1/2]_0$ laser transitions in atomic xenon at wavelengths 1.73, 2.03 and 2.65 μm, respectively.

The emission parameters of these lasers have been studied in many papers. However fundamental data on the kinetics and spectroscopic parameters of the corresponding transitions are still insufficient. Thus, the probabilities (oscillator strengths) of the $5d \rightarrow 6p$ transitions have been calculated in papers [3, 4], and the total lifetime of the $5d[3/2]_1$ level in pure xenon was measured in [5] by exciting the $5d[3/2]_1$ level by a laser pulse and studying its radiative relaxation in the VUV range to the ground state.

At the same time, the data on the laser transition probability, population processes and the lifetime of the upper level of the active medium are very important because they determine the cross section for stimulated emission, the efficiency and 'accumulation ability' of the laser, i.e. the possibility of its operation in the master oscillator – amplifier scheme.

It seems that experimental studies in this field have been impeded so far by the absence of sufficiently sensitive and fast IR photodetectors required for studying the time dependence of the luminescence intensity at laser transitions in xenon.

Recently, certain progress has been achieved in the development of such detectors. In particular, PD 24-03 and PD 36-03 photodiodes have been fabricated, which can be used in such experiments. The aim of our paper is to measure the time dependence of the spontaneous emission intensity at the $5d[3/2]_1 \rightarrow 6p[3/2]_1$ and $5d[3/2]_1 \rightarrow 6p[5/2]_2$ transitions at 2.03 and 1.73 μm, respectively, upon excitation of pure xenon by an electron beam of short duration for obtaining the data on the population and relaxation kinetics of the $5d[3/2]_1$ level of the Xe atom.

2. Experimental

Figure 1 shows the scheme of the experimental setup. The active medium was excited by a Radan-220 pulsed electron accelerator [6] emitting 150-keV, 2-ns electron pulses with the current amplitude of ~ 500 A. The accelerator operated in the single pulse regime. A cell was a quartz tube with internal diameter 24 mm and length 150 mm and the output optical BaF_2 window. The internal surface of the

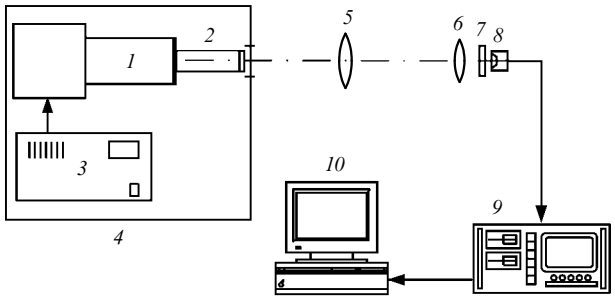

Figure 1. Scheme of the experimental setup: (*1*) Radan-220 electron accelerator; (*2*) cell; (*3*) accelerator power supply; (*4*) magnetically soft iron housing; (*5*) lens with the focal distance $F = 170$ mm and diameter 80 mm; (6) lens with $F = 40$ mm and diameter 40 mm; (*7*) IKS-1500 optical filter; (*8*) PD 24-03 photodiode; (*9*) Tektronix TDS 1012 storage oscilloscope; (10) personal computer.

I.A. Denezhkin, P.P. D'yachenko State Scientific Center of Russian Federation, A.I. Leipunskii Institute of Physics and Power Engineering, pl. Bondarenko 1, 249033 Obninsk, Kaluga region, Russia; e-mail: denezhkin@ippe.ru; dpp@ippe.ru

Received 2 June 2008; revision received 10 September 2008
Kvantovaya Elektronika **39** (2) 135 – 138 (2009)
Translated by M.N. Sapozhnikov

tube was covered with a mirror aluminium coating. The cell was evacuated with a backing pump through a liquid-nitrogen trap down to a pressure of 5×10^{-3} Torr and was filled with a medium under study.

The influence of electromagnetic interference produced by the accelerator on the operation of a detector was reduced by two methods. First, the power supply and accelerator with the cell fixed on it were placed inside a housing made of magnetically soft iron. One of the walls of the housing opposite to the cell window had a hole of diameter 30 mm to extract optical radiation. Second, a photodetector was located at a distance of ~ 1 m from the accelerator. The emission from the cell was focused on the photodetector by using a special optical system. The output window of the cell was imaged with a long-focus quartz lens with aperture $\varnothing$80 mm at a distance of ~ 80 cm and then was projected with a short-focus quartz length through an IKS-1500 optical filter on the entrance pupil of the photodetector.

The detector was an PD 24-03 photodiode with the entrance pupil and photosensitive area diameters of 7 and 0.3 mm, respectively. The detector operated in the photovoltaic regime (with a load of 50 Ω). The output signal of the detector, corresponding to a luminescence pulse (spontaneous emission) was fed to a Tektronix TDS 1012 storage oscilloscope connected with a PC.

A number of auxiliary and control measurements were performed before the main measurements. First, we studied the spectral response function of the detector system. Figure 2 shows the relative spectral sensitivity of the PD 24-03 photodiode [7] and the transmission spectrum of an IKS-1500 filter recorded with a Cary-500 spectrophotometer. One can see that the system is sensitive in the wavelength range from 1.3 to 2.5 μm. The photosensitivity of the system outside this range is close to zero. Measurements performed with an MDR-23 monochromator (with a 300-lines mm^{-1} diffraction grating and an PD 24-03 photodiode) showed that pure xenon excited by an electron beam emits in this spectral range only lines at 2.03 and 1.73 μm, corresponding to the $5d[3/2]_1 \rightarrow 6p[3/2]_1$ and $5d[3/2]_1 \rightarrow 6p[5/2]_2$ transitions, respectively. The intensity of the first line is approximately an order of magnitude higher than that of the second one. The detection of the total emission in these lines enhances the effective light-gathering power of the spectrometer approximately by 10 %. Such a detection does not distort the measurements of excitation and relaxation of the $5d[3/2]_1$ level because these transitions share the same upper level.

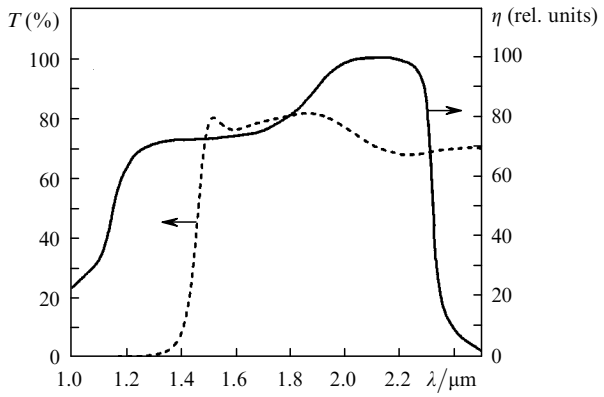

Figure 2. Wavelength dependences of the spectral sensitivity η of a PD 24-03 photodiode and transmission spectrum T of an IKS-1500 optical filter.

Second, we verified the linearity of the spectrometer with the help of attenuating filters. A light source was a working cell with pure xenon at a pressure of 20 Torr excited by an electron beam. The operation stability of the accelerator was controlled by detecting visible radiation with a surface-barrier silicon photodetector [8]. The spectrometer was found to be linear within ±10 % in the pulse amplitude range from 0.5 to 30 mV used in experiments.

We also measured the temporal response function of the spectrometer. A single 2-ns pulse from a G5-59 generator (the pulse shape was controlled with a C-75 oscilloscope) was fed to a detector and recorded with a Tektronix TDS 1012 oscilloscope operating in the same regime as upon detecting the emission of xenon. The pulse FWHM was 4 ± 0.5 ns, corresponding to the time constant $RC \sim 3$ ns of the system, which is consistent for the load 50 Ω with the certificated values of the capacities of the PD 24-03 photodiode and a connecting cable equal approximately to 40 and 20 pF, respectively.

We found also that the possible presence of quenching impurities in the medium under study does not affect experimental results. For this purpose, we compared the amplitude and half-width of luminescence pulses under different experimental conditions, in particular, by using vessels with spectrometrically pure xenon obtained from a production plant at different times, working cells made of different materials (quartz, stainless steel) and also after different hold times of the medium in the cell (up to 16 hours). No noticeable differences in the pulse shape were found within the experimental error. Nevertheless, during measurements at different xenon pressures, the cell was evacuated each time and filled with fresh gas.

The experimental method is described in more detail in [9].

3. Experimental results and discussion

The experimental results are presented in Figs 3–5. Oscillations observed at the trailing edge of luminescence pulses in Fig. 3 are caused by the electromagnetic interference produced by the accelerator. The errors shown in Figs 4 and 5 are obtained from the scatter of the results of a series of measurements and characterise mainly the reproducibility of the parameters of a pulse of the electron accelerator.

The shape of the luminescence pulse weakly depends on the xenon pressure and the rise and decay times of the pulse, for example, at a pressure of 1 Torr are 3 and 10 ns, respectively (see Fig. 3a). It follows from this, without a detailed analysis of the pulse shape, that the lifetime of the $5d[3/2]_1$ level does not exceed 10 ns. This approximately an order of magnitude shorter than the lifetime observed upon excitation of the $5d[3/2]_1$ level by a laser pulse [5].

Such a considerable difference between the lifetimes of the $5d[3/2]_1$ level in these two cases can be explained by the fact that upon excitation of xenon by an electron beam, unlike excitation by a laser pulse, this level is efficiently quenched by electrons of a recombining plasma. This confirms a hypothesis [10] about electron mixing of laser levels explaining the delay of the Ar–Xe laser pulse observed upon electron-beam pumping.

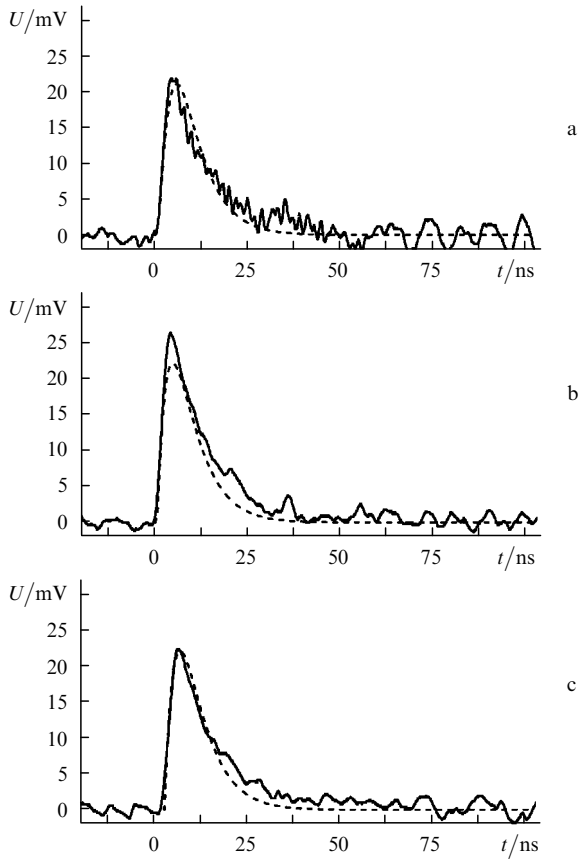

Figure 3. Luminescence pulses of pure xenon excited by an electron beam at pressures $p = 1$ (a), 16 (b) and 70 Torr (c). Solid curves: experiment; dashed curves: calculation.

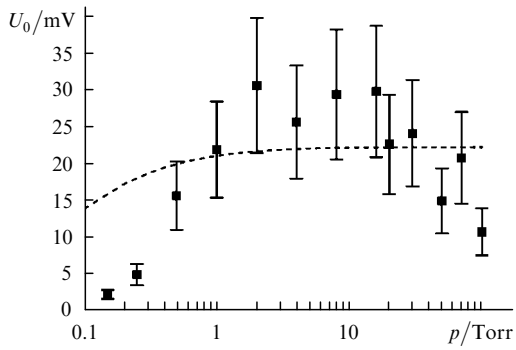

Figure 4. Dependence of the luminescence pulse amplitude U_0 on the xenon pressure p. Squares: experiment; the dashed curve: calculation.

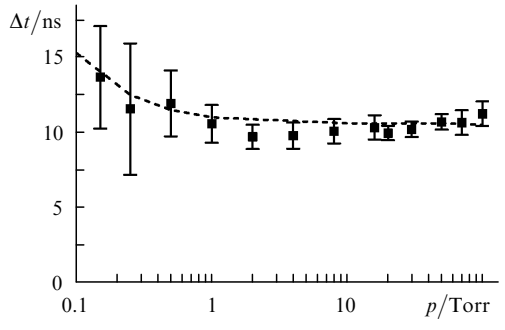

Figure 5. Dependence of the luminescence pulse half-width Δt on the xenon pressure p. Squares: experiment; the dashed curve: calculation.

At the same time, the dependences of the amplitude and half-width of the luminescence pulse on the gas pressure are somewhat unexpected at a glance. In particular, the fact that these parameters almost do not change with increasing the xenon pressure almost by two orders of magnitude. However, taking into account the finite duration of the accelerator pulse and the time resolution of the spectrometer, these experimental dependences can be explained within the framework of a rather simple model.

It is known that the population $N(t)$ of the $5d[3/2]_1$ level during its excitation and relaxation can be described by the equation

$$\frac{dN(t)}{dt} = R(t) - \frac{N(t)}{\tau}, \tag{1}$$

where $R(t)$ and τ are the excitation rate and lifetime of this level, respectively. The excitation rate $R(t)$ and voltage pulse $U(t)$, which is proportional to the spontaneous emission intensity $Q(t)$, can be written in the form

$$R(t) = A_p \exp\left(-\frac{t}{\tau_p}\right), \tag{2}$$

$$U(t) = kQ(t) = k\,\frac{N(t)}{\tau_{rad}}, \tag{3}$$

where A_p and τ_p are the pump rate at the initial moment and its relaxation time constant; $\tau_{rad}^{-1} = \tau_1^{-1} + \tau_2^{-1}$; τ_1 and τ_2 are the radiative time constants for the $5d[3/2]_1 \rightarrow 6p[5/2]_2$ transition at 1.73 μm and the $5d[3/2]_1 \rightarrow 6p[3/2]_1$ transition at 2.03 μm, respectively; k is the calibration constant connecting oscilloscope readings with the spontaneous emission intensity. By substituting expressions for $R(t)$ and $U(t)$ into (1), we obtain

$$\frac{dU(t)}{dt} = \frac{k}{\tau_{rad}}\,A_p \exp\left(-\frac{t}{\tau_p}\right) - \frac{U(t)}{\tau}. \tag{4}$$

The solution of this equation, taking into account the accelerator pulse duration τ_{acc} and the time resolution τ_0 of the spectrometer, has the form

$$U(t) = k\,\frac{A_p}{\tau_{rad}}\,\frac{\tau_p}{\tau_p - \tau_{acc}}\left\{\frac{\tau\tau_p}{\tau_p - \tau}\left[\frac{\tau_p}{\tau_p - \tau_0}\left(e^{-t/\tau_p} - e^{-t/\tau_0}\right)\right.\right.$$

$$\left. - \frac{\tau}{\tau - \tau_0}\left(e^{-t/\tau} - e^{-t/\tau_0}\right)\right] - \frac{\tau\tau_{acc}}{\tau_{acc} - \tau}\left[\frac{\tau_{acc}}{\tau_{acc} - \tau_0}\right.$$

$$\left.\left. \times\left(e^{-t/\tau_{acc}} - e^{-t/\tau_0}\right) - \frac{\tau}{\tau - \tau_0}\left(e^{-t/\tau} - e^{-t/\tau_0}\right)\right]\right\}. \tag{5}$$

The dashed curves in Figs 3–5 are calculated by (5) by using the best-fit parameters $\tau_{acc} = 3$ ns, $\tau_0 = 6$ ns, $\tau_p^{-1} = 4 \times 10^9 p$, $\tau^{-1} = 0.9 \times 10^9 p^{1/2}$, and $A_p = 1.1 \times 10^4 p^{3/2}$, where τ_p and τ are expressed in seconds, A_p in $cm^3\ s^{-1}$, and the xenon pressure in torr. One can see that the calculated curves are consistent as a whole with experimental data, except the regions of very low ($p < 0.5$ Torr) and large ($p > 70$ Torr) pressures in Fig. 4. This can be explained by the fact that the given model neglects the resonance nature of the $5d[3/2]_1$ state of the Xe atom, which is manifested at low pressures, and the saturation of the energy input of the electron beam into the medium at high xenon pressures.

Note that the optimal time resolution of the spectrometer proved to be $\tau_0 = 6$ ns rather than 3 ns, as in the control experiment with the use of the generator. This is explained by the fact that in the latter case the charge accumulation time in a photodiode, which was probably ~ 5 ns in our case, was neglected.

The power dependence of the parameters τ and A_p on the xenon pressure is explained by the fact that the electron density in the recombining plasma increases with pressure as $p^{1/2}$ [10].

Thus, we can say that the population and relaxation of the $5d[3/2]_1$ level upon excitation of pure xenon by an electron beam is mainly determined by the excitation and quenching of this level by electrons of the recombining plasma, the time constants of these processes being very small. Thus, at xenon pressures of a few torr used in the laser, the lifetime of the $5d[3/2]_1$ level at least in the time interval comparable with the pump pulse duration is almost completely determined by electron quenching and is ~ 1 ns.

At the same time, it can be shown that the electron density required to produce efficient quenching with the rate constant, for example, of 2×10^{-7} cm^3 s^{-1} [10] considerably exceeds the estimate of the density based on the calculation of the energy losses of the electron beam in xenon. Because these losses in the given case are small, we can assume that a considerable increase in the electron density under our experimental conditions can be caused, in particular, by the secondary electron emission from cell walls.

Note also that luminescence pulses (Fig. 3) are well described by the model proposed above and do not contain long-lived components. This means that excitation of pure xenon does not initiate plasma-chemical population of the $5d[3/2]_1$ level. In particular, the level population proceeding via the formation of molecular Xe_2^+ ions does not occur. This result is of interest because the dissociative recombination of the Xe_2^+ ion is considered in papers devoted to the kinetics of processes in pure xenon [11, 12] as the main pumping channel of a xenon laser.

4. Conclusions

We have measured the time dependences of the spontaneous emission intensity at the $5d[3/2]_1 \rightarrow 6p[3/2]_1$ and $5d[3/2]_1 \rightarrow 6p[5/2]_2$ transitions at 2.03 and 1.73 μm, respectively, upon excitation of pure xenon by a short-pulse electron beam in a broad pressure range. Analysis of our results has shown that the population and relaxation of the $5d[3/2]_1$ level are mainly determined by its excitation and quenching by electrons of the recombining plasma. The characteristic time constants of these processes are very small and do not exceed 2 ns. Thus, at pressures of a few torr used in a Ar–Xe laser, the lifetime of the $5d[3/2]_1$ level, at least in the interval comparable with the electron pulse duration, is almost completely determined by electron quenching and is ~ 1 ns.

Upon excitation of pure xenon by an electron beam, no plasma-chemical processes of population of the $5d[3/2]_1$ level have been found. In particular, population via the formation of molecular Xe_2^+ ions, which is considered as an important pumping channel of a xenon laser, is absent.

Acknowledgements. The authors thank V.P. Semenov for his help in the study and useful discussions. This work was supported by Rosatom Contract No. N.4d.47.03.08.123.

References

1. Karelin A.V., Sinyavskii A.A., Yakovlenko S.I. *Kvantovaya Elektron.*, **24**, 387 (1997) [*Quantum Electron.*, **27**, 375 (1997)].
2. Gulevich A.V., D'yachenko P.P., Zrodnikov A.V., Kononov V.N. *Atom. Energy*, **80**, 361 (1996).
3. Aymar M., Coulombe V. *Atomic Data and Nuclear Data Tables*, **21**, 537 (1978).
4. Dasgupta A., Apruzese J.P., Zatsarinny O., Bartschat K., Fischer C.F. *Phys. Rev. A*, **74**, 012509 (2006).
5. Alekseev V.A., Setser D.W. *J. Phys. Chem. A*, **103**, 8396 (1999).
6. Baksht E.Kh., Kostyrya I.D., Lipatov E.I., Lomaev M.I., Rybka D.V., Tarasenko V.F. *Zh. Tekh. Fiz.*, **77**, 98 (2007).
7. Denezhkin I.A., D'yachenko P.P. Preprint FEI-3057 (Obninsk, 2005).
8. Lapidus V.I., D'yachenko P.P., Seregina E.A., Semenova N.N. *Prib. Tekh. Eksp.*, (3), 108 (2001).
9. Denezhkin I.A., D'yachenko P.P. Preprint FEI-3124 (Obninsk, 2008).
10. Peters P.J., Yun Fu Lan, Ohwa M., Kushner M.J. *IEEE J. Quantum Electron.*, **26**, 1964 (1990).
11. Karelin A.V., Simakova O.V. *Kvantovaya Elektron.*, **28**, 121 (1999).
12. Dudin A.Yu., Semenova L.V., Ustinovskii N.N., Kholin I.V., Chugunov A.Yu. *J. Sov. Laser Res.*, **13**, 374 (1992).