Time behaviour of the 7^3S and 6^3P_1 sates after narrow band laser excitation of the 6^3P_1 state of mercury

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ABSTRACT

Investigations of the behaviour of mercury transitions are reasonable task for improvement of different types light sources as well as for solving environmental problems. Recently, the time behaviour of the 546.0 nm spectral line was studied after two step pulse laser excitation of the 7^3S state via the 6^3P_1 state.[1] A nonmonotonical decay of the population of 7^3S was observed and was interpreted as a result of the stimulated emission in forward direction of the excitation laser beam.

In our experiment we observe the nonmonotonical behaviour of the decay of the 546.0 nm spectral line intensity when the 6^3P_1 state is excited by strong pulse laser beam. This behaviour is changing when the wavelength of excitation laser beam is tuned around the 253.6 nm wavelength.

Some hypothesis for explanation of this behaviour are given.

1. EXPERIMENT

The experimental set-up is presented in Figure 1. The quartz cell was mounted in a oven, which temperature was controlled with an accuracy of 2^0C. The cell temperature was kept approximately 50^0C higher than the temperature of a reservoir with natural isotope mixture of mercury. This reservoir had a separate heating. Excitation of the mercury atoms in the cell was carried out by tuneable laser radiation in the UV region.

The laser system consisted of a Nd:YAG pumped dye laser (Spectra Physics, Quanta Ray GCR 170, Nd:YAG; Radiant Dyes DL midi E dye laser), operated with Cumarin 30 solved in Isopropanol. The needed wavelength around 253.6 nm was obtained by doubling of the dye laser wavelength by an angle - tuned BBO nonlinear

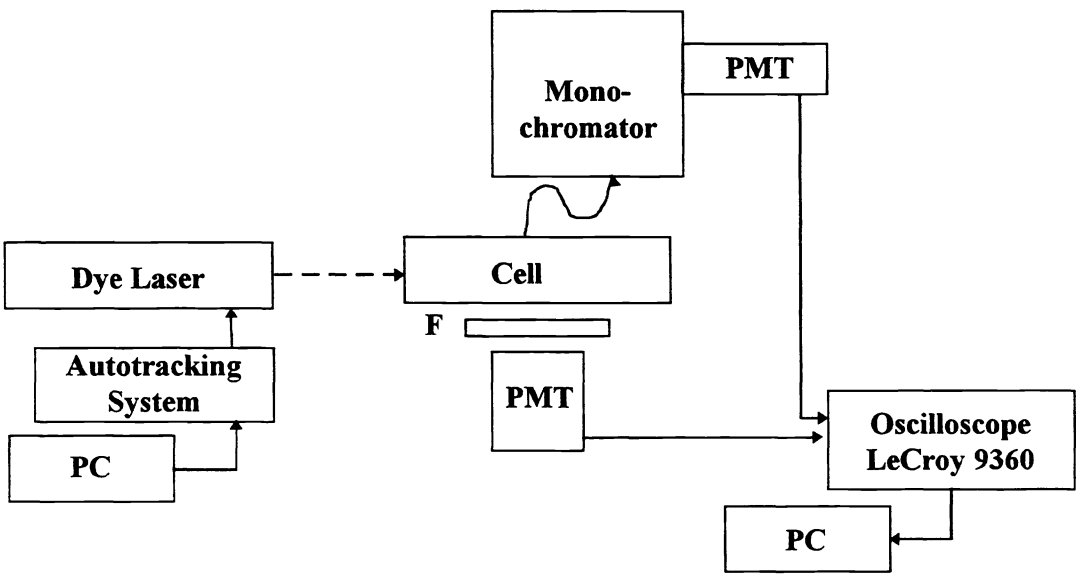

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Figure 1. Experimental set - up. Monochromator channel is used for 253.6 nm spectral line registration.
crystal. The UV radiation was separated from the fundamental laser wavelength by four Pelen - Broka prisms. While scanning the dye laser wavelength, the power of the UV radiation was kept within the limit of ± 10% by a computer controlled autotracking system. The UV radiation power was 1.5 mJ and the laser beam was 2 mm in diameter in the excitation region. The wavelength resolution of the laser system in the region of 253.6 nm was 0.001 nm. The scanning was made step by step. Each step was 0.005 nm, which is not the limit of the set-up. According to the atom mercury concentration and the Doppler width between 15 and 27 steps were necessary to scan the laser frequency around the mercury intercombination line. The 546.0 nm spectral line was separated by a interference filter, while the fluorescence at 253.6 nm was separated by a grating monochromator. Both spectral lines were detected by fast photomultipliers Hamamatsu R - 955 type. The signals were averaged using a digital oscilloscope (LeCroy - 9360) over 1000 laser pulses.

2. RESULTS

In Figures 2 the time behaviour of the 546.0 nm (7^3S_1 - 6^3P_1) spectral line is presented.

In the first part of the curve, the laser excitation pulse and the 7^3S_1 effective lifetime dominate the shape. Practically, they are masked in the increasing part of the curve. This part is followed by a nonmonotonical part, which shows two maxima.

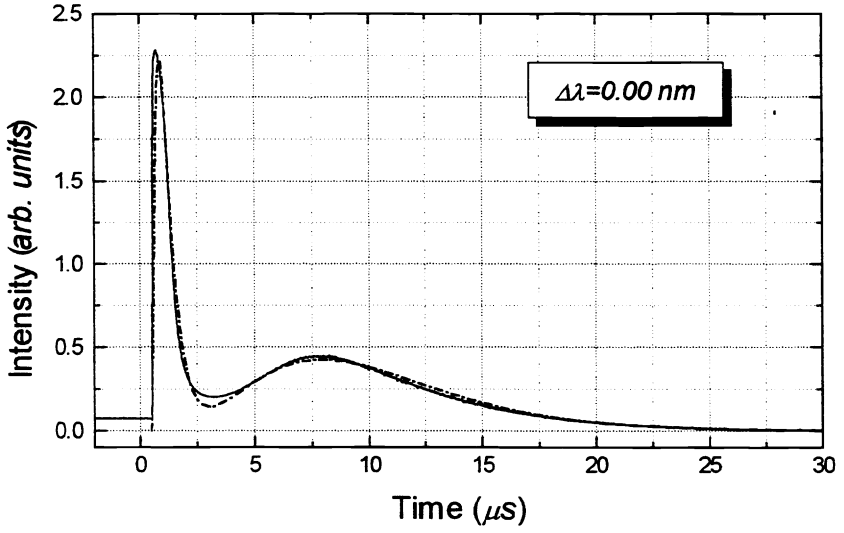

Figure 2. Time behaviour of 546.0 nm spectral line (solid line - experiment; dash line - simulation at zero laser wavelength detuning).

The first maximum has a fast decay, while the second maximum has a long tail.

When the wavelength of the excitation laser beam is changed from maximal red detuning to shorter wavelengths a second maximum appears close to the first one. This maximum, further moves to larger delay times, when the excitation laser wavelength approach the centre of the 253.6 nm spectral line.

The maximum value and the maximum value of the delay time of the second maximum is observed before the wavelength of the excitation beam approaches the centre of the 253.6 nm spectral line. The time behaviour is reproduced mainly, when the wavelength of the excitation beam is detuned from the centre of the 253.6 nm spectral line to the shorter wavelengths. This behaviour is presented on Figure 3.

This behaviour was observed at different density of the mercury atoms. The concentration is varied in the interval from 0.3 Torr to 9.6 Torr.

The time dependence of the second maximum position as well as its value vs detuning of laser wavelength is presented in Figure 4.

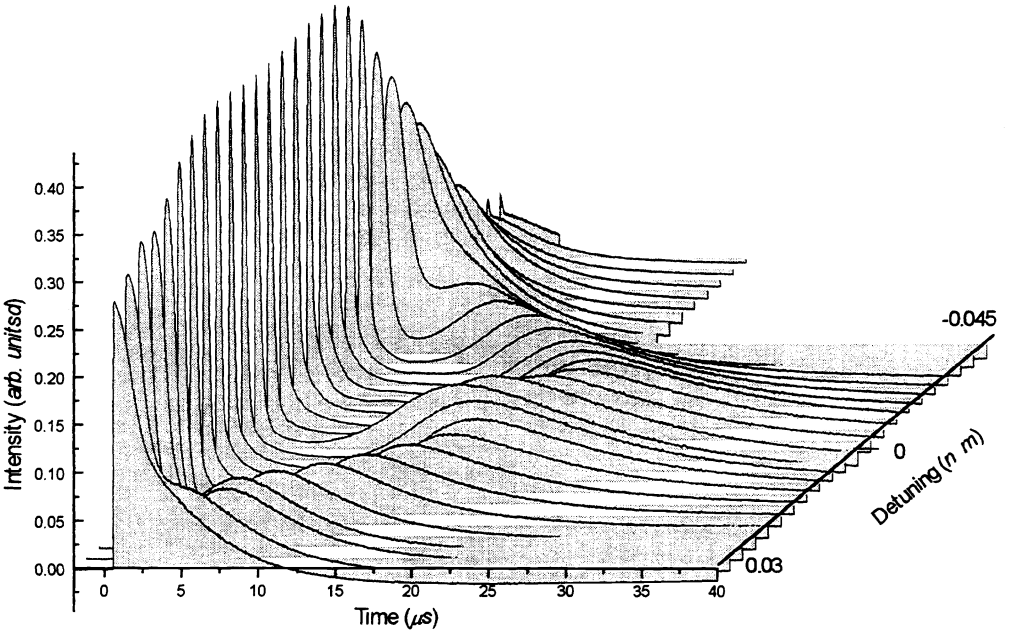

Figure 3. Evolution of the 546.0 nm decay curve vs laser wavelength detuning around 253.6 nm.

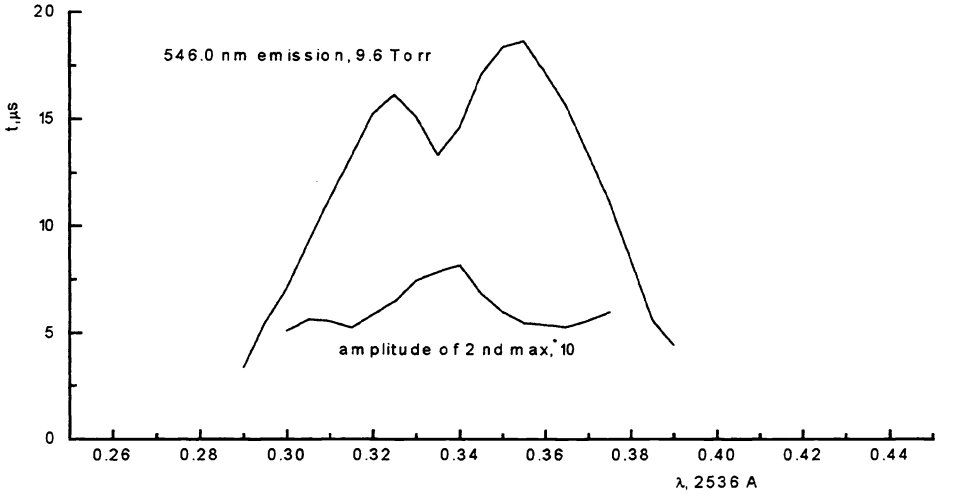

Figure 4. Position and amplitude(arb. units) of the 546.0 nm spectral line second maximum vs excitation laser wavelength.

In the experiment, simultaneously the time behaviour of intercombination line 253.6 nm was recorded. The decay has a long tail, which decay time does not change while tuning the laser beam wavelength, and a small maximum appear, which coincides with the first maximum of 546.0 nm spectral line decay curves. The second 546.0 nm spectral line maximum reproduces itself on the 253.6 nm curve in a smaller extend.

3. DISCUSSION

It is clear that in this experiment the population of the 7^3S_1 state is determined by collision processes (Figure 5). After pulse excitation of the 6^3P_1 state, due to collisions of excited 6^3P_1 atoms the high lying states of Hg are populated and the fluorescence spectrum of Hg exhibits many lines from these states. Hence, the population can be ascribed partially to Energy Poling (EP) processes.[2,3] The cascade transitions from these states to 7^3S state, EP and radiative trapping form the first maximum of the 546.0 nm emission. In this way all 6^3P states are populated via the famous green - blue triplet. Further, the collisions between all 6^3P atoms, mainly Associative Ionisation(AI):

$$Hg(6^3P_0) + Hg(6^3P_1) \rightarrow Hg_2^+ + e^- \quad (1)$$

and Penning Ionisation (PI):

$$Hg(6^3P_2) + Hg(6^3P_2) \rightarrow Hg^+ + Hg_0 + e^- \quad (2)$$

followed by Dissociate Recombination(DR) and population of high lying states provide the second maximum. These processes have previously been shown to be very efficient.[4]

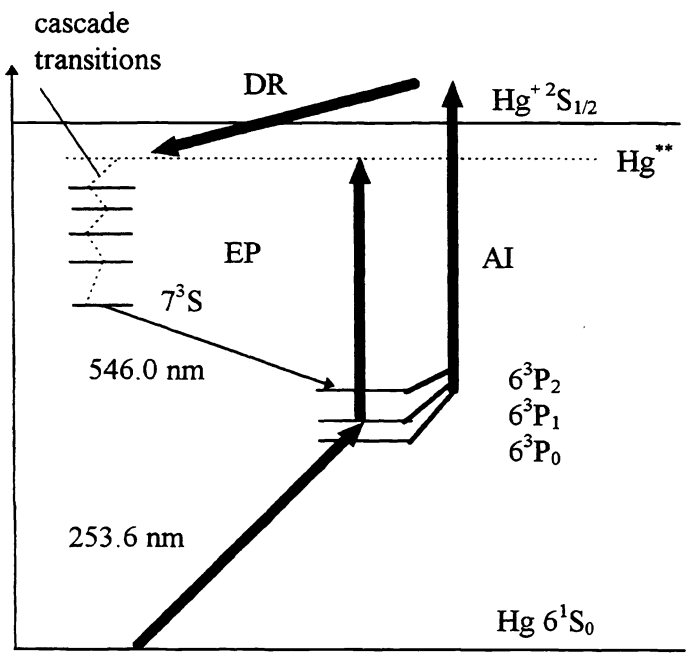

Figure 5 Simplified energy level diagram of HgI representing the relevant processes for our investigation.

These processes seems to be the driving mechanism leading to the non-monotonic time behaviour of the 546.0 nm spectral line and the long time delay observed in our experiment.

There are several reasons, which could cause the time evolution of the observed signals with detuning of the excitation laser wavelength.

- A simulation was done, taking into account the processes mentioned above and radiation trapping of all states, considered in the model, in which 5 arbitrary intermediate states are assumed. This model explain the fast part of the decay curves. Employing this model, we also can explain the variation of the time evolution with the detuning of the excitation laser around the line centre(Figure 3). The decrease of the delay time of the second maximum at resonant excitation of the 6^3P_1 state we ascribe to absorption at the line centre. For the relative population of the 6^3P_1 states this means, that there exist a situation similar to detuning of the laser wavelength, as we observe in the line wing. The dash line in Figure 2 presents this model results.
- In this experiment, groups of atoms, having different velocities are exited when the excitation laser wavelength is changed from the 253.6 nm wing to the line centre. It is obvious, that atoms having larger energies approach smaller

internuclear distances of quasimolecule, which is created in the (1) (2) processes. It is well known that at smaller internuclear distances transition probabilities of quasimolecule decay are larger, so the DR process could be realised at shorter delay times, which we have observed in the experiment.

- The last speculation is connected with the fact that in the cell illuminated by a strong laser field many electrons are created by Penning and Associative ionisation. These electrons are, further, involved in the Dissociate and Tree particle recombinations. The cross sections of these processes increase dramatically with electron temperature decreasing. On the other hand, these electrons are thermalised via second kind collisions with ground state atoms.

In our experiment, when the laser wavelength is changed from the wing of the 253.6 nm spectral line to the centre, the number of the atoms, which are excited to the 6^3P_1 state, i. e. which leave mercury ground state are increasing and could reach value more than 50 % of the total atom concentration. Hence, the ground state atoms, which are involved in the electron thermalisation decrease. So, the time of the thermalisation increases and the corresponding cross sections reach their maximum values at larger delay times.

All these hypothesis are possible and corresponding processes could play simultaneously role in the present experiment.

4 CONCLUSION

In this paper, the time behaviour of the 546.0 nm spectral line is investigated after strong laser excitation of the 6^3P_1 state. This behaviour changes when the excitation laser wavelength is tuned around the 253.6 nm spectral line profile. Energy poling, Associative ionisation and Dissociate recombination are proposed as driving processes. Some hypothesis are proposed for the background of the observed signals.

5. ACKNOWLEDGEMENT

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6. REFERENCES

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