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# Radiative-lifetime and Landé-factor measurements of the Se I $4p^3 5s\ ^5S_2$ level using pulsed laser spectroscopy

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**Abstract.** — First laser spectroscopic investigations on atomic selenium are reported. Natural selenium was thermally dissociated in a quartz resonance cell keeping the background pressure of abundant selenium molecules low by differential heating. The $4p^3 5s\ ^5S_2$ level was excited by frequency-tripled pulsed-dye laser radiation at 207 nm. From time-resolved recordings of the fluorescence decay at 216 nm a natural radiative lifetime of 493(15) ns was determined, while quantum-beat and optical double-resonance measurements in an external magnetic field yielded $g_J = 2.0004(10)$ for the Landé factor. The results are compared with previous theoretical and experimental data.

## 1. Introduction.

With the development of semiconductor technology and xerography, selenium has found wide applications. Selenium is also widely known in biology and agriculture as a trace element. Some compounds of selenium are very toxic. Thus it is important to be able to detect small amounts of selenium from a technological as well as from an ecological point of view. It is also of interest to determine the abundance of selenium in astronomical objects. Presently the abundance of selenium in the sun is unknown [1]. In order to assess selenium concentrations in remote emission measurements it is necessary to know transition probabilities of suitable selenium spectral lines. Comparatively little information about transition probabilities and associated radiative lifetimes exists for selenium. Studies of the selenium atom are complicated, since the resonance lines fall in the far ultra-violet and vacuum-ultraviolet spectral regions and since selenium vaporizes as different kinds of polyatomic molecules. Actually, no previous measurements with selective excitation methods exist.

In recent years it has become easy to generate powerful short-wavelength laser radiation employing pulsed laser sources. E.g., in our laboratory several studies of lifetimes and hyperfine structures with excitation around 200 nm have been performed using time-resolved spectroscopy as well as level-crossing and optical double-resonance spectroscopy adopted to

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pulsed excitation (See, e.g. [2-4]). In the present work we report on lifetime and $g_J$ factor measurements for the $4p^3 5s\,^5S_2$ level of selenium, that can be reached by 207 nm laser excitation. The corresponding state ($5p^3 6s\,^5S_2$) of the heavier Group VIA atom tellurium was investigated by resonance spectroscopy following rf-lamp excitation 20 years ago [5]. Selenium is more difficult to investigate since the atomic fraction in the vapor is smaller than for tellurium. However, with laser excitation this did not prove to present a problem. The selenium and tellurium quintet states can only decay to the ground configuration $^3P$ states in transitions that are partly allowed because of the break-down of LS coupling. Since this coupling in more pure for the lighter element selenium, the quintet state lifetime is expected to be considerably longer than for tellurium ($\tau(\mathrm{Te}\ 5p^3 6s\,^5S_2) = 71.8(2.2)$ ns [5]). Clearly, measurements of the Landé factor will also yield information on the coupling conditions.

A number of theoretical calculations of the selenium quintet-state lifetime exist, that, however, exhibit large internal deviations. Lawrence [6] and Gruzdev [7] have used a semi-empirical method using the Coulomb approximation for intermediate coupling states. Garpman *et al.* [8] performed relativistic self-consistent-field calculations using the dipole-velocity and dipole-length operators. No direct measurements of the quintet level lifetime exist. Beam-foil data exist for the $^3S_1$ level of the same configuration [9]. Using this information combined with emission and absorption measurements of relative transition probabilities Berzinsh *et al.* [10, 11] could estimate the quintet state lifetime to 290(100) ns. This is in strong contrast to the only other experimental estimate based on the 207 nm selenium line oscillator strength obtained in emission measurements by Corliss and Bozman [12]. In view of this situation an experiment aiming at an accurate determination of the lifetime and the $g_J$ factor was performed using time-resolved and optical double resonance (ODR) laser spectroscopy on selenium vapor in a sealed-off quartz resonance cell. Our experimental set-up is described in the next section. In section 3 our measurements are reported and in a final section the results are discussed.

## 2. Experimental set-up.

The experimental set-up used in the present experiments is shown in figure 1. High-temperature selenium resonance cells were prepared specially for this experiment following the procedure used in reference [5]. A cell consisted of a cylinder, 30 mm in diameter and 40 mm long, to which a 16 cm long stem is attached. The cells were baked out at 750 °C in high vacuum for about 15 hours prior to distilling a small amount of elemental selenium into the cell and sealing off. Natural selenium, consisting of 92 % even-even isotopes ($A$ = 74, 76, 78, 80, and 82) and 8 % of $^{77}$Se (nuclear spin $I$ = 1/2) was used for the cells. During the experiments the cells were placed in a differentially heated oven, allowing the cell cylinder to be kept at 100-200 °C higher temperature than the stem. The coldest point of the cell determines the total (mainly molecular) vapor pressure in the cell, while the hotter cell temperature determined the degree of molecular dissociation into free atoms. Temperatures were measured with thermocouples. The selenium vapor pressure was inferred from reference [13].

The oven was made from non-magnetic materials (including the heating wire, that was made out of platinum) and was placed in Helmholtz coil systems producing a well-defined magnetic field. The rest of the experimental set-up is similar to the one described in reference [3]. About 2 mJ of pulsed laser radiation at 207 nm was generated by frequency tripling the output from a Nd : YAG-laser-pumped dye laser. Fluorescence light on the 216 nm selenium line was isolated with a grating monochromator and was detected by a Hamamatsu R331 photomultiplier tube. In the lifetime measurements and the quantum-beat

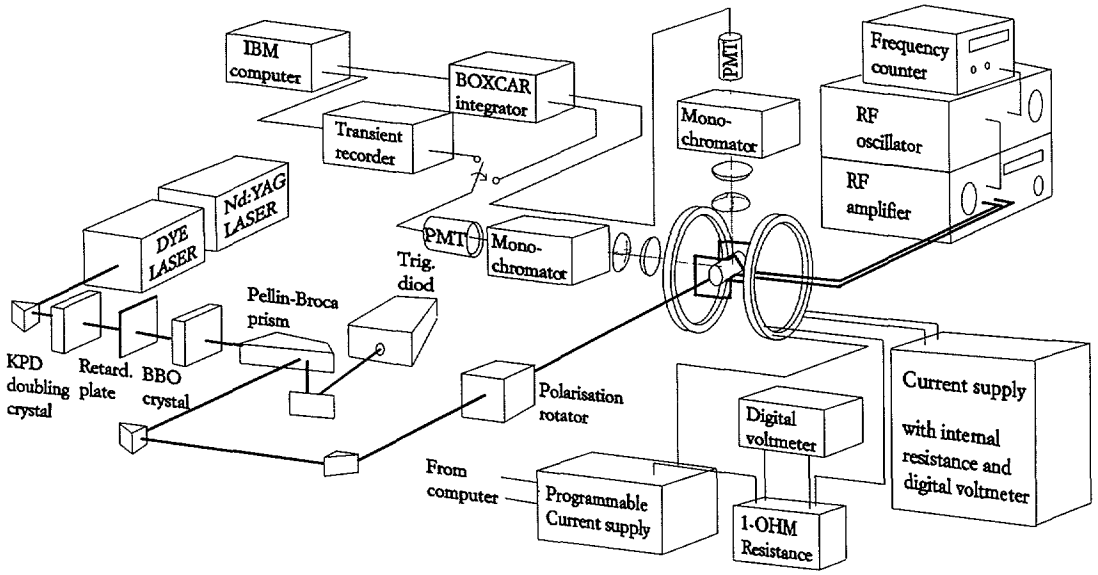


Fig. 1. — Experimental set-up used in laser spectroscopy on selenium atoms.

experiments, the signals were captured and averaged with a Tektronix Model DSA 602 transient digitizer and were transferred to an IBM-compatible computer for evaluation.

In the ODR experiment, two pairs of Helmholtz coils were used to produce the magnetic field. One coil system was utilized to produce a static offset field from which a periodic sweep field could be generated with the other coil system for the ODR measurements. Rf transitions were induced by a two-turn coil connected to the rf equipment. The output of a General Radio 1215-C unit oscillator was amplified in a Boonton Radio 230A tuned amplifier. In the ODR experiments a Stanford Research SR 265 boxcar integrator connected to an IBM-compatible AT computer was used for data collection and processing. Because of rather large pulse-to-pulse fluctuations in the excitation source, the fluorescence was detected in two directions and the signals were divided to improve the signal-to-noise ratio [3].

### 3. Measurements and results.

Lifetime measurements were performed in a magnetic field strong enough to wash out any quantum-beat signals, that could otherwise cause deviations from a pure exponential decay of the excited state. Series of recordings were taken for decreasing cell temperatures and the evaluated inverse lifetime (the decay constant) was plotted as a function of the total selenium pressure as shown in figure 2. The plots were found to be linear over three orders of magnitude of vapor pressure. The extrapolated value for zero density yields the lifetime, that was found to be

$$\tau(4p^3 5s\ ^5S_2) = 493(15)\ \text{ns}\ .$$

The uncertainty in the value reflects the statistical scatter in the data and is also somewhat influenced by uncertainties in the temperature measurements. We note, that the measured lifetime is quite long. Considering that the measurements were performed in a sealed-off cell without possibilities of further evacuation, the question arises whether the value might be

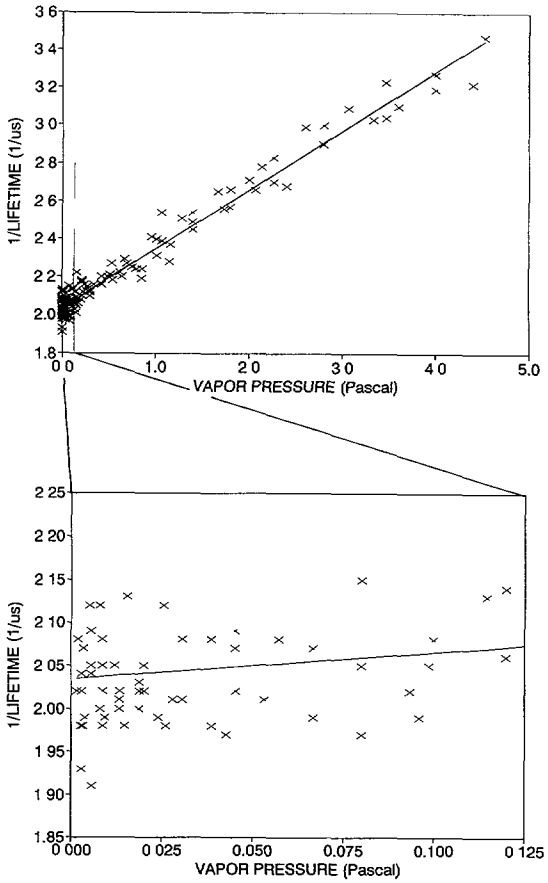


Fig. 2. — Measured decay constant for the $4p^3 5s\ ^5S_2$ level of selenium as a function of selenium vapor pressure.

shortened by collisions with residual rest gas present also in the cold cell. We have specially addressed this question. First we note, that the same lifetime value was obtained for two different cells, prepared at the same time. Even after artificial aging of one of the cells by heating at 600 °C for about 5 hours the lifetime was unaffected. No tendency of flattening out for low temperatures is observed in plots such as the one in figure 2. We measured the decay times for different combinations of linearly polarized light for excitation and detection including « magic angle » (54.7°) arrangement. Extrapolations to zero selenium vapor pressure yielded the same lifetime independent of polarizer settings.

The Landé $g_J$-factor was measured by observing $\Delta m = 2$ Zeeman quantum beats, and by optical double-resonance experiments. All measurements were performed in well-defined external magnetic field. The magnetic field was accurately calibrated in separate experiments, by observing optical pumping signals in the ground state of $^{133}Cs$.

An example of a quantum-beat recording is shown in figure 3 together with a Fourier transform yielding the beat frequency. We were not able to obtain the desired accuracy in the quantum-beat measurements, since the time response of our set-up was not fast enough to allow measurements in high fields. A more precise determination of the Landé $g_J$-factor was made in optical double-resonance measurements. Using $\pi$ excitation ($\Delta m_J = 0$), the $^5S_2$ level was populated and the fluorescence light emitted from the transition to the

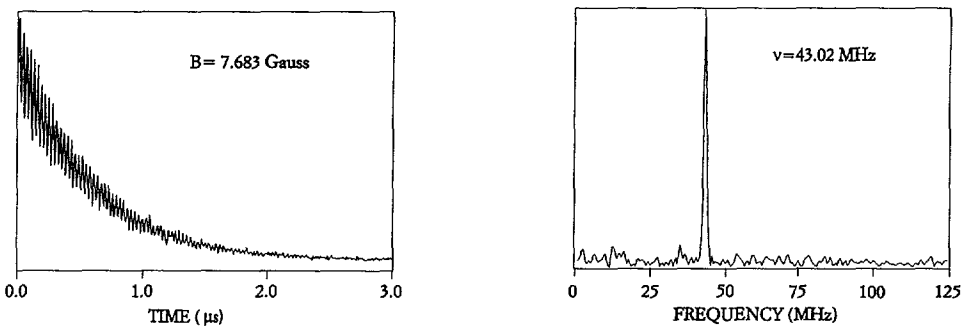


Fig. 3. — Zeeman quantum-beat recording and a calculated Fourier-transform spectrum for the $4p^3 5s\ ^5S_2$ level of selenium.

$^3P_1$ level was detected. When resonant magnetic dipole transitions ($\Delta m_J = \pm 1$) were induced by the rf field a decrease in $\sigma$ light was obtained. Using a second detector, a combination of $\pi$ and $\sigma$ radiation with an intensity unaffected by the rf transitions was detected to provide signal normalization. For practical reasons, the magnetic field was swept through resonance rather then sweeping the radio frequency. In figure 4 an ODR signal is shown. From the ODR measurements and Zeeman quantum-beat experiments we obtain

$$g_J(4p^3 5s\ ^5S_2) = 2.0004(10)\,.$$

The error is determined by statistical scattering and by a magnetic-field calibration uncertainty of 2 parts in $10^4$.

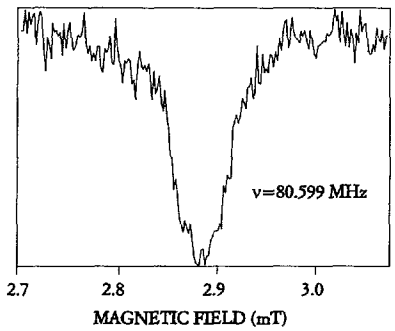


Fig. 4. — ODR signal for the $4p^3 5s\ ^5S_2$ level of selenium.

## 4. Discussion.

Our experimentally determined lifetime value for the $4p^3 5s\ ^5S_2$ selenium state is compared with the available literature data in table I. The best agreement with our data is obtained for the relativistic dipole-length calculations by Garpman *et al.* [8]. Actually, by neglecting relativistic effects the same authors obtained the lifetime value 1 300 ns using the same operator. The non-relativistic calculations presented in references [6, 7] also yield values of the lifetime that are longer than those obtained from Garpman's relativistic calculation.

Table I. — *Radiative lifetimes for the* $4p^3 5s\ ^5S_2$ *level of selenium.*

| Reference | | Lifetime [ns] |
|---|---|---|
| *Experimental :* | | |
| This work | | 493(15) |
| Berzinsh *et al.* [10, 11] | | 290(100) |
| Corliss and Bozman [12] | | < 89 |
| *Theoretical :* | | |
| Lawrence [6] | | 1 128 |
| Gruzdev [7] | | 2 390 |
| Garpman *et al.* [8] | DL | 440 |
| | DV | 960 |

There is also a deviation between our result and the value given by Berzinsh and Ubelis [10, 11]. As mentioned above, their value strongly relies of the beam-foil result by Dynefors [9] for the $^3S_1$ state ($\tau$ = 1.7(0.2) ns). We are planning a laser spectroscopic re-measurement for this very short-lived state, for which a different experimental approach must be taken.

Using the intermediate wave functions given by Gruzdev [7] and Garpman [8] a $g_J$ value of 1.996 is calculated for the $4p^3 5s\ ^5S_2$ state. Our experimental value 2.0004 is found to be closer to the pure LS limit 2.0023.

It should also be noted that all quoted theoretical data are semi-empirical in nature relying on experimentally determined energy-level positions to calculate the degree of intermediate coupling. Since these calculations were performed more accurate energy-level information for the selenium atom has become available [14-16].

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This paper is dedicated to Professor Pierre Jacquinot, celebrating a most remarkable scientific career, extending over 60 years.

**References**

[1] GREVESSE N., *Phys. Scr.* **T 8** (1984) 49.
[2] BENGTSSON J., LARSSON J., SVANBERG S. and WAHLSTROM C.-G., *Phys. Rev. A* **41** (1990) 23.
[3] BENGTSSON J., LARSSON J. and SVANBERG S., *Phys. Rev. A* **42** (1990) 5457.
[4] BENGTSSON G. J., LARSSON J., SVANBERG S. and WANG D. D., *Phys. Rev. A* **45** (1992).
[5] GARPMAN S., LIDO G., RYDBERG S. and SVANBERG S., *Z. Physik* **247** (1971) 238.
[6] LAWRENCE J. M., *Astrophys. J.* **148** (1967) 261.
[7] GRUZDEV P. F., *Opt. Spectr.* **27** (1969) 479.
[8] GARPMAN S., HOLMGREN L. and ROSÉN A., *Phys. Scr.* **10** (1974) 221.
[9] DYNEFORS B. J., *Phys. Scr.* **11** (1975) 375.
[10] BERZINSH U., Ph. D. Thesis, University of Latvia (Riga, 1988).
[11] UBELIS A. and BERZINSH U., *Phys. Scr.* **34** (1986) 805.

[12] CORLISS C. H. and BOZMAN W., Experimental Transition Probabilities for Spectral Lines of Seventy Elements, N.B.S. Washington D.C. (1962).
[13] VON ARDENNE M., Tabellen zur Angewandten Physik, Part 2 Berlin (1964).
[14] ERIKSSON K. B. S., *Phys. Lett.* **41** A (1972) 97.
[15] MORILLON C. and VERGES J., *Phys. Scr.* **10** (1974) 227.
[16] LINDGREN B. and PALENIUS P., *Sol. Phys.* **53** (1977) 347.