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Oscillator strength of the strongly "forbidden" Pb $6p^2\ ^3P_0 \rightarrow 6p^2\ ^3P_1$ transition at 1278.9 nm

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Abstract. The "forbidden" Pb $6p^2\ ^3P_0 \rightarrow 6p^2\ ^3P_1$ line at 1278.9 nm is measured by diode laser absorption in a resistively heated hot-pipe filled with a lead vapour and argon as buffer gas. The measurements performed at a temperature of 1170 K and a lead number density of 2.4×10^{15} cm^{-3} yield the oscillator strength $f_{\rm F} = (4.5 \pm 1.1) \times 10^{-7}$ which corresponds to a radiative transition probability $A_{\rm F} = (6.1 \pm 1.5)$ s^{-1}. Within the error bars, the result is in agreement with theoretical data published by different authors.

PACS. 32.70.-n Intensities and shapes of atomic spectral lines – 32.70.Cs Oscillator strengths, lifetimes, transition moments

For many years the spontaneous emission rates for multipole lines belonging to the transitions between the lowest lying lead $6p^2$ levels (see Fig. 1) have been subject of calculations using different theoretical approaches [1–5]. The calculations yielded diverse values of the radiative transition rates for the $6p^2\ ^1S_0 \rightarrow 6p^2\ ^3P_J$ and $6p^2\ ^1D_2 \rightarrow 6p^2\ ^3P_J$ transitions, while the results reported for the near IR strongly forbidden transitions $6p^2\ ^3P_0 \rightarrow 6p^2\ ^3P_1$ (1278.9 nm) and $6p^2\ ^3P_0 \rightarrow 6p^2\ ^3P_2$ (938.9 nm) between the ground and the lowest lying metastable states, are in good mutual agreement. For instance, the values for the 1278.9 nm line, recently reported by Biemont and Quinat [4] and Horodecki *et al.* [5] are very close (within 5%) to the first theoretical result ($A_{\rm F} = 7.14$ s^{-1}) obtained by Garstang [1].

Experimental data available in literature for the spontaneous emission rates of the considered multipole lines are scant. The measurements of the relative intensities of the forbidden Pb lines corresponding to the $6p^2\ ^1S_0 \rightarrow 6p^2\ ^3P_J$ and $6p^2\ ^1D_2 \rightarrow 6p^2\ ^3P_J$ transitions [6–8] yielded the ratios of the spontaneous emission rates for these transitions. The ratios of the calculated transition probabilities given by Garstang [1] differ form the values following from the measurements [6,7], while the latest results published by Horodecki *et al.* [5] agree well with the experiment. To our best knowledge, experimental data for the spontaneous emission rates for the 938.9 nm and 1278.9 nm lines have not yet been reported. The transition at 1278.9 nm has been investigated experimentally, but all experiments that have been reported so far [9–12], deal with the investigation of the parity non-conserving (PNC) optical rotation in atoms. The most recent measurements performed by Meekhof *et al.* [10] and Phipp *et al.* [12] reported the value of the ratio $R = {\rm Im}\langle E1^{\rm PNC}\rangle / \langle M1\rangle$ ($E1$ and $M1$ are PNC and magnetic dipole amplitude, respectively). However, the experiments of Meekhof *et al.* [10] and Phipp *et al.* [12] have not yielded the information about the spontaneous emission rate $A_{\rm F}$, since their absorption measurements which provided only the value of the product $N\langle M1\rangle^2 \sim NA_{\rm F}$, were short of determining the atom number density N.

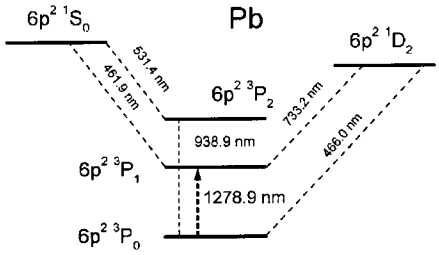

Fig. 1. Partial term diagram displaying the ground $6p^2$ configuration of neutral lead with multipole transitions indicated.

Here, we report the results of the first experimental determination of the oscillator strength and the corresponding spontaneous emission rate for the led 1278.9 nm line. The measurements were performed in a stainless-steel hot-pipe with quartz windows at the ends and filled with argon as a buffer gas. The central part of the oven, containing a high-purity natural abundant lead (^{208}Pb: 52%, ^{207}Pb: 22%, ^{206}Pb: 25%, ^{204}Pb: ≈1%), was resistively heated. The beam of a single-mode DFB diode laser (Laser Components, type SPECDILAS D-Series, line width: 10 MHz, side mode suppression: 25 dB) was transmitted through the hot-pipe oven and absorption signals were detected

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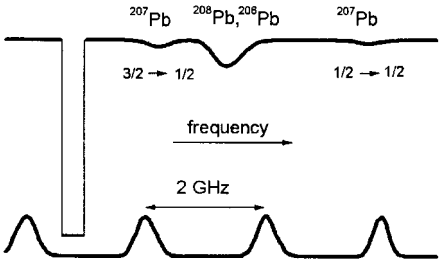

Fig. 2. Measured absorption spectrum recorded by scanning the laser across the $6p^2\ ^3P_0 \rightarrow 6p^2\ ^3P_1$ transition (upper trace) with frequency dispersion marks of the Fabry-Perot interferometer (lower trace). For the ^{207}Pb lines the values $F \rightarrow F'$ are indicated.

by a near-infrared InGaAs PIN photodiode (Hamamatsu). The power of the laser beam (1 mW) was reduced by several density filters to avoid saturation of the photodetector. The wavelength of the laser was scanned back and forth by current at a fixed temperature. A part of the laser intensity reflected by a glass plate in the optical path was used to obtain frequency marks from a 2 GHz confocal Fabry-Perot interferometer. The frequency sweep was wider than shown in Figure 2 (typically 12 GHz) and small distortion of the scan linearity could be observed only near the turning points where the sweep reverses direction. Pb densities were chosen to obtain optically thin conditions and a maximum peak absorption of 10–20%.

A typical spectrum is displayed in Figure 2 together with the Fabry-Perot interferometer dispersion marks. The central component is slightly broader than the side ones and has an asymmetric shape. It represents the unresolved $6p^2\ ^3P_0 \rightarrow 6p^2\ ^3P_1$ transitions in ^{208}Pb, ^{206}Pb and the weak isotope ^{204}Pb. The other two components are the hyperfine transitions in ^{207}Pb. The nuclear spin of the ^{207}Pb isotope $I = 1/2$, so that the $6p^2\ ^3P_1$ level in the ^{207}Pb splits in two components with total angular momentum $F = 3/2$ and $F = 1/2$. The energy defects of the hyperfine $^3P_1$ sublevels with total angular momentum $F = 3/2$ and $F = 1/2$ are $(1/2)A_{207}(^3P_1)$ and $(-1)A_{207}(^3P_1)$, respectively, where $A_{207}(^3P_1)$ is the hyperfine splitting parameter. The relative intensities for the hyperfine components $F = 3/2 \rightarrow F = 1/2$ and $F = 1/2 \rightarrow F = 1/2$ within the $^3P_1 \rightarrow ^3P_0$ transition are 2:1. Apparently (see Fig. 2) the value for $A_{207}(^3P_1)$ is negative. The dipole hyperfine splitting parameter $A_{207}(^3P_1)$ evaluated from the obtained spectrum amounts to (-2390 ± 30) MHz. This value is in very good agreement with the result previously reported by Reeves and Fortson [13] who found $A_{207}(^3P_1) = (-2388 \pm 4.5)$ MHz. The agreement can serve as an additional proof of the adequate laser scan linearity in the present experiment.

The oscillator strength f_F of the measured forbidden line can be determined using the expression

$$W_F^\nu = \frac{\pi e^2}{mc} N_{Pb} L f_F, \qquad (1)$$

which is valid for optically thin lines. Here, W_F^ν is the line equivalent width expressed in frequency units, N_{Pb} is the lead number density and L is the effective vapour column length. In the present experiment the density of the vapour column was not perfectly homogeneous. The effective column length $L = (4 \pm 0.5)$ cm was determined from the length of lead deposition on a very thin tungsten wire placed in the hot-pipe axis. In the first, straightforward evaluation step we determined the temperature *via* the Doppler width taken from the strongest ^{207}Pb hyperfine component. The line widths were measured at various argon pressures in the range between 5 and 50 mbar. The equivalent width $W_F^\nu = 1.15 \times 10^8$ s^{-1} was independent of the noble gas pressure, *i.e.* constant within an error bar of $\pm 5\%$. In the zero pressure limit the Doppler width was found to be $(2/\lambda)(2 \ln 2kT/M)^{1/2} = (400 \pm 10)$ MHz, which yielded $T = 1190 \pm 50$ K. According to the lead vapour pressure curve given by Nesmeyanov [14], the corresponding lead number density was 1.5×10^{15} cm^{-3} $< N_{Pb} < 6 \times 10^{15}$ cm^{-3}. Taking into account the Pb number density and the length of the absorbing layer the oscillator strength f_F calculated according to equation (1) is in the range $1.8 \times 10^{-7} < f_F < 7.2 \times 10^{-7}$. The corresponding radiative transition probability is 2.4 s^{-1} $< A_F <$ 9.8 s^{-1}.

In order to improve the accuracy of f_F, the temperature and lead number density were determined by an additional measurement of the optically thick lead resonance line at 283.4 nm ($6p^2\ ^3P^0 \rightarrow 7s\ ^3P_1^0$ transition). The resonance absorption was measured using the continuum of a deuterium lamp and a 0.5 m Jarell-Ash monochromator (spectral resolution: 0.03 nm) with an RCA 1P28 photomultiplier. The measured equivalent widths were more than two orders of magnitude larger than the corresponding Doppler width and strongly dependent on noble gas pressure. In the applied pressure range the resonance line wings were of the Lorentzian shape, essentially due to noble gas broadening and, in addition, due to self-broadening. At argon pressures higher than 100 mbar the resonance line exhibited quasistatic broadening in the red wing. The curve of growth of the resonance line, *i.e.* the behaviour of the equivalent width W_R^ν is given by [15]

$$(W_R^\nu)^2 = \frac{2\pi e^2}{mc}(N_{Ar}\gamma_{Ar}^\nu + N_{Pb}\gamma_{Pb}^\nu) N_{Pb} L f_R. \qquad (2)$$

Here, N_{Ar} is argon number density, γ_{Ar}^ν and γ_{Pb}^ν are the coefficients for impact broadening due to argon and lead, respectively, and f_R is the resonance line oscillator strength. In the evaluation we used the value $f_R = (0.212 \pm 0.003)$ given by Penkin and Slavenas [16]. As predicted by relation (2), $(W_R^\nu)^2$ exhibited linear dependence on the argon pressure in the applied range (5–50 mbar). A fit through the measured $(W_R^\nu)^2$ data (see Fig. 3) was a straight-line with the slope $\Delta(W_R^\nu)^2/\Delta p_{Ar} = (5.6 \pm 0.5) \times 10^{20}$ s^{-2} mbar^{-1}. The straight-line intercepted the ordinate axis at $(w_R^\nu)^2 = (2 \pm 1) \times 10^{21}$ s^{-2}. This residuum should be attributed to the self-broadening contribution in equation (2). Applying Dalton's law, one can rewrite expression (2) in the following form:

$$N_{Pb} = \frac{mc}{2\pi e^2}\left[(W_R^\nu)^2 - (w_R^\nu)^2\right] \frac{kT}{p_{Ar}} \frac{1}{\gamma_{Ar}^\nu L f_R}. \qquad (3)$$

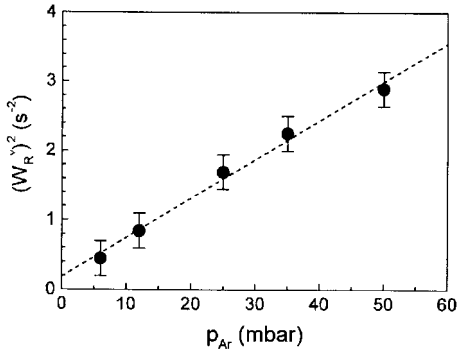

Fig. 3. The dependence of the square of the equivalent width of the led resonance line on the argon pressure. The dashed line represents the least square fit through the measured data.

Here, it was assumed that $N_{\mathrm{Ar}} \gg N_{\mathrm{Pb}}$, *i.e.* that the measured pressure is equal to the argon partial pressure. Equation (3) can be regarded as valid with an accuracy of about 3% for our experimental conditions. On the other hand, combination of Dalton's law and the lead vapour pressure curve given by Nesmeyanov [14] shows that in the temperature range 1000–1200 K the equilibrium lead number density as a function of temperature can be adequately described by:

$$N_{\mathrm{Pb}} = 10^{a-b/T} \tag{4}$$

where the coefficients a and b are (23.185 ± 0.025) and (9160 ± 25) K^{-1}, respectively. Taking into account the values measured for $(W_{\mathrm{R}}^{\nu})^2$ in dependence on argon pressure and $\gamma_{\mathrm{Ar}}^{\nu} = (8.5 \pm 0.5) \times 10^{-10}$ $\mathrm{cm}^3\,\mathrm{s}^{-1}$ reported by Franzke *et al.* (2000), the following solutions to the system of equations (3, 4) in terms of lead number density and temperature were found: $N_{\mathrm{Pb}} = (2.4 \pm 0.9) \times 10^{15}$ cm^{-3}; $T = (1170 \pm 25)$ K. The declared error bars include the uncertainties of the vapour pressure curve (±10%), the experimental errors of the equivalent widths (±5%), the broadening coefficient and the uncertainty of the vapour column length.

The oscillator strength can be determined using a combination of equations (1, 3) which yields:

$$f_{\mathrm{F}} = 2\,\frac{p_{\mathrm{Ar}}}{kT}\gamma_{\mathrm{Ar}}^{\nu}\frac{W_{\mathrm{F}}^{\nu}}{[(W_{\mathrm{R}}^{\nu})^2 - w_{\mathrm{R}}^{\nu})^2]}f_{\mathrm{R}}. \tag{5}$$

The evaluation of relation (5) for a series of the measured $(W_{\mathrm{R}}^{\nu})^2$ *versus* p_{Ar} values yields:

$$f_{\mathrm{F}} = 4.5 \times 10^{-7}$$

and the corresponding

$$A_{\mathrm{F}} = 6.1\ \mathrm{s}^{-1}$$

with a statistical accuracy of about 10%. Taking into account the errors of the broadening coefficient and the oscillator strength of the resonance line, and the equivalent width of the forbidden line, the overall uncertainties of the f_{F} and A_{F} amount to 25%.

Table 1. Values for the radiative transition probability A_{F} of the multipole lead line at 1278.9 nm. MCDF − multiconfiguration Dirac-Fock, MCRRPA − multiconfiguration relativistic random-phase approximation, HFR − relativistic Hartree-Fock, ICC − intermediate coupling calculations.

	$A_{\mathrm{F}}(\mathrm{s}^{-1})$	Method
Theory	7.27	MCDF (experimental energy values)[a]
	4.29	MCDF (theoretical energy values)[a]
	7.50	Semiempirical[a]
	7.85	HFR[b]
	7.00	MCRRPA[c]
	7.08	HFR[d]
	7.14	ICC[e]
Experiment	6.1 ± 1.5	Laser absorption[f]

[a] Reference [5], [b] reference [4], [c] reference [3], [d] reference [2], [e] reference [1], [f] this work.

In Table 1 our experimental value for A_{F} is compared with theoretical data obtained by other authors. Considering the experimental error bar, our result is in agreement with the theoretical values.

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