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Measurements of the oscillator strengths for the 6p7s $(1/2,1/2)_1 \rightarrow 6pnp\ (1/2,3/2)_2$ Rydberg transitions of lead

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ABSTRACT

We report experimentally determined oscillator strength distribution in the bound region of lead corresponding to the 6p7s $(1/2, 1/2)_1 \rightarrow 6pnp\ (1/2, 3/2)_2$ $(20 \leq n \leq 52)$ Rydberg transitions. The absolute value of the photoionization cross section from the 6p7s $(1/2, 1/2)_1$ excited state at the first ionization threshold 6p $(^2P_{1/2})$ has been determined as 27 ± 4 Mb using the saturation absorption technique. The threshold value of the photoionization cross section is used to calibrate the oscillator strengths of the above mentioned Rydberg transitions. Moreover, oscillator strengths in the bound region smoothly connect with the differential oscillator strength density at the first ionization threshold, and verify the fundamental condition of quantum defect theory.

1. Introduction

The knowledge of oscillator strength is vital for the determination of chemical abundance of different elements in the sun and stars. Therefore, various models have been developed to measure the abundance of lead in the atmospheres, interior of stars, and in the solar atmosphere. The earliest available data on the oscillator strengths of lead were mainly acquired to determine its abundance in the sun and stars. The solar spectrum shows six prominent lines of lead associated with the $6p^2\ ^3P_{0,\,1,\,2}, ^1D_2 \rightarrow 6p7s\ ^3P^0_{0,\,1,\,2}$ transitions at 283.3, 363.9, 405.7, 722.9, 368.3 and 374.0 nm. The determination of the solar abundance of lead has attracted much attention of scientists during the last few decades to exploit different theoretical and experimental techniques [1–5]. DeZafra and Marshall [6] reported the oscillator strengths from the $^3P_1$ excited state of Pb from the lifetimes and branching ratios for the three important decay channels from the $^3P_1$ state of lead. Later, Grevesse [7] summarized the oscillator strengths of the above mentioned transitions of lead reported by different groups to determine the abundance of lead in the sun. Ganas [8] used the analytical independent particle model potential to calculate the *f*-values of the $6p^2\ ^3P_0 \rightarrow 6p\ ns\ ^3P_1$ $(7 \leq n \leq 10)$ and $6p^2\ ^3P_0 \rightarrow 6pnd\ ^3D_1$ $(6 \leq n \leq 8)$ transitions and compared his results with the previously reported theoretical and experimental data. Recently, Bieron et al. [9] computed the oscillator strengths for the $np^2 \rightarrow (n+1)s$ transitions of lead using the relativistic configuration approach and compared the results with the earlier data. Sansonetti and Martin [10] compiled the data of transition probabilities (A_{ki}) of various elements including lead in a Handbook of Basic Atomic Spectroscopy Data by NIST. Vadla et al. [11] reported the oscillator strength of the strongly forbidden $6p^2\ ^3P_0 \rightarrow 6p^2\ ^3P_1$ transition of lead at 1278.9 nm.

The above literature survey shows that, there is no experimental or theoretical data available on the oscillator strengths of the 6p7s $(1/2, 1/2)_1 \rightarrow 6pnp\ (1/2, 3/2)_2$ Rydberg transitions of lead. In the present contribution, we present the absolute value of the photoionization cross section from the 6p7s $(1/2, 1/2)_1$ intermediate state at the first ionization threshold of lead using the saturation absorption technique and subsequently used this value to determine the oscillator strengths of the 6p7s $(1/2, 1/2)_1 \rightarrow 6pnp\ (1/2, 3/2)_2$ $(20 \leq n \leq 52)$ Rydberg transitions.

2. Experimental details

Our laser system is an injection seeded Q-switch Nd:YAG laser (Spectra Physics) having 7–9 ns pulse duration and 30 Hz repetition rate. A two-step laser excitation scheme for the investigation of the Rydberg states and for the measurement of photoionization cross section is shown in Fig. 1. The first dye laser, operated with R-610 dye and pumped with the second harmonic (532 nm) was frequency doubled and fixed at 283.3 nm to populate the 6p7s $(1/2, 1/2)_1$ intermediate state from the $6p^2\ (1/2, 1/2)_0$ ground state. The second dye laser was operated with a mixture of Stilbene-420 and LD-390 dyes and was pumped with the third harmonic (355 nm) of the same Nd:YAG laser to cover the energy range from 59325 to 59850 cm^{-1} up to the first ionization limit. Both the exciting and scanning dye laser beams were inserted into the thermionic diode ion detector from opposite sides and their spatial overlap was ensured in the interaction region. The dye lasers were Hanna type [12] having spectral width $\leq 0.3\ cm^{-1}$. The vapor containment system is a thermionic diode ion detector, 35 cm long and 4 cm internal diameter,

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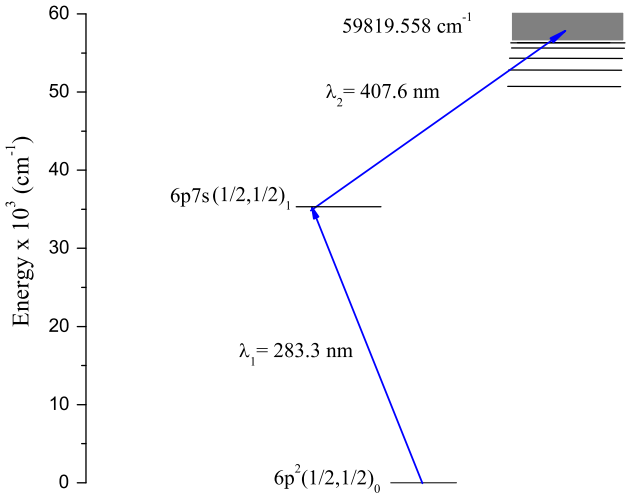

Fig. 1. Two-step laser excitation scheme for the investigation of Rydberg states and photoionization cross section measurement of lead.

operating in the space charge limited mode. The detector was evacuated to 10^{-6} mbar using a combination of rotary and diffusion pump. About 2–3 g of lead was loaded in a stainless steel boat and placed in the middle of the detector which was heated up to 1170 K using a thermo-coax wire and temperature was monitored through a Ni–Cr–Ni thermocouple. The heat pipe was filled with argon gas at a pressure of ≈2–4 mbar to reduce the vapor diffusion and to protect the windows. The temperature profile of the central heating region was monitored by placing thermocouples along the heat pipe and was found to be almost uniform at 1170 K. This uniform temperature distribution and constant argon pressure ensured the uniform column density in the central heating zone. A 0.25 mm thick molybdenum wire, stretched axially along the heat pipe and heated by a regulated power supply, was used as a cathode. The change in the diode current due to the photoion production was measured as a voltage drop across a 100 kΩ load resistor. The linearity of the detection system was verified on the strongest photoionization signal; therefore the amplitude of the photoionization signal was proportional to the number of ions produced during the laser pulse. For the wavelength calibration, a small fraction of the scanning laser beam was fed into a neon filled hollow cathode to record the optogalvanic spectrum. For the relative wavelength markers, another fraction of the beam was passed through a 1 mm thick solid etalon (3.33 cm^{-1} free spectral range), and its transmission fringes were recorded. All the three signals from the thermionic diode detector, the hollow cathode lamp and the photo-diode were processed with three box-car averagers (SR 250) and saved on a computer through a GPIB (NI488) interfacing card for subsequent analysis. The energies of the unknown lines of the Rydberg series were analyzed through a computer program which takes into account the reference signals of neon and used the etalon rings to interpolate between the lines. The uncertainties associated with the determination of the term energies are ± 0.25 cm^{-1}. The Rydberg states have been recorded using the above mentioned procedure and in the following section the procedure for the measurement of photoionization cross section of the excited state and oscillator strengths of Rydberg transitions has been described.

3. Results and discussion

The optical oscillator strengths of the 6p7s $(1/2, 1/2)_1 \rightarrow 6pnp$ $(1/2, 3/2)_2$ Rydberg transitions of lead have been determined using the experimental technique developed by Mende and Kock [13]. This technique has been extensively used for the determination of oscillator strengths of Rydberg transitions of alkali metals, alkaline earths and group-IIB elements by Baig and coworkers [14–19]. The technique is applicable for the measurement of oscillator strengths for medium to high principal quantum numbers n, with the assumption that ionization probability of the transitions that lie within K_BT approaches unity (K_B is the Boltzmann constant and T is temperature in Kelvin). As long as the condition of the ionization probability is fulfilled there is a well established relation between the f-values and the photoionization cross-section at the first ionization threshold:

$$f_n = \frac{4\pi\varepsilon_o mc}{\pi e^2}\left(\frac{S^{Ryd}}{S^+}\right)\left(\frac{\lambda^+}{\lambda^n}\right)\sigma\left(\lambda^+\right). \quad (1)$$

Here f_n is the oscillator strength of the nth transition of Rydberg series, that is directly proportional to the photoionization cross-section σ measured at threshold wavelength λ^+. The {S^{ + }} is the ion signal at the ionization threshold (see Fig. 3) and S^{Ryd} is the integrated ion signal intensity for the nth transition at a wavelength λ^n. The integrated signal intensity is equal to the peak value times the half-width, that is $S^{Ryd} = Ic\Delta k$, where I is the signal intensity, c is the speed of light and k is the wave number in cm^{-1}. The constants e, m, c and ε_o are the charge of electron, the mass of the electron, speed of light and the permittivity of free space respectively.

It is obvious from the above relation that the value of the photoionization cross section at threshold is an important parameter for the determination of the oscillator strengths of the Rydberg transitions. Therefore, we have first measured the photoionization cross section from the 6p7s $(1/2, 1/2)_1$ intermediate state of lead at the first ionization threshold using the saturation absorption technique as describe by Burkhardt et al. [20] and He et al. [21]. This technique is based on the two-step ionization process, in the first step the atoms are excited from the ground state to an intermediate state and the intensity of the first laser (exciting) is kept sufficiently high to saturate the excited state. The second laser (ionizing) is fixed at a wavelength corresponding to the ionization threshold and its intensity is varied using neutral density filters and at each insertion corresponding amplitude of the photoionization signals are registered on a storage oscilloscope and the pulse energy of the ionizing dye laser is measured by an energy meter.

The relation for the measurement of photoionization cross section can be derived for the two-step photoionization process including the saturation phenomena. In this method, the high intensity laser pulse tends to equalize the population of the two levels, thus the saturation is expressed as:

$$N_1(t) = N_2(t) = \frac{N_T(t)}{2}. \quad (2)$$

Here, $N_1(t)$ and $N_2(t)$ are the number densities of atoms in the ground and excited state at time t respectively, and $N_T = N_1(t) + N_2(t)$. The change in the number density of the excited states due to the photoionization can be written as:

$$\frac{dN_T(t)}{dt} = -\frac{I(t)\sigma}{h\nu}N_2(t) = -\frac{I(t)\sigma}{2h\nu}N_T(t). \quad (3)$$

Here $I(t)$ is the laser intensity at a certain wavelength and σ is the photoionization cross section of the intermediate state. Rearranging and taking integral of the above equation gives:

$$\int_{N_0}^{N_T}\frac{dN_T(t)}{N_T(t)} = -\frac{\sigma}{2h\nu}\int_0^t I\left(t'\right)dt' \quad (4)$$

where $N_T(t) = N_o$ at $t = 0$. Integrating and simplifying the above equation:

$$N_T = N_o e^{-\sigma/h\nu\int_o^t I(t')dt'}. \quad (5)$$