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Two-step photoionisation of palladium

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***Abstract*. The efficiency of two-step ionisation of palladium involving the 276.4-nm $4d^{10}\,{}^1S_0 - 4d^9 5p\,{}^3P_1$ transition is measured as a function of the laser radiation intensity in experiments aimed at laser separation of isotopes. It is shown that to obtain the efficient (over 50 %) two-step ionisation of palladium by 10-ns laser pulses with a pulse repetition rate of 10 kHz, an average radiation intensity should exceed 100 W cm^{-2}. The results of comparison of theoretical and experimental data on the two-step ionisation efficiency make it possible to estimate the photoionisation cross section of the $4d^9 5p\,{}^3P_1$ state ($\sigma_i \approx 10^{-16}$ cm^2).**

Keywords: *laser isotope separation, two-step photoionisation, photoionisation cross section.*

1. Introduction

The prospects of laser isotope separation (LIS) for obtaining the rare isotope ^{168}Yb in grams was demonstrated in papers [1 – 4]. The results of theoretical and experimental investigations were summarised in reviews [5 – 7] (see, for example, Refs [8, 9] for a description of the earlier stages of LIS investigations). It seems quite natural to try to use the experience gained during the production of ^{168}Yb to obtain other rare isotopes required in medicinal applications. The isotope ^{102}Pd is one of the commercially prospective materials of this kind.

In order to determine the possibility of commercial production of an isotope by the LIS technique, it is extremely important not only to know the isotopic structure of the selective transition, but also to have information about the cross section of the photoionisation transition because this cross section determines the required power of the ionising laser radiation. The possibility of selective photoionisation of palladium was discussed in Refs [10, 11]. The two-step ionisation of odd isotopes of Pd using radiation from a single laser at 276 nm was studied in Ref. [10]. In this work, we consider the two-photon ionisation of palladium using a single laser. The results of experimental and theoretical investigations which can be used for determining the photoionisation cross section are presented. These results differ considerably from the data presented in Ref. [10].

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2. Photoionisation scheme of ^{102}Pd

The ground state of palladium is $4d^{10}\,{}^1S_0$ (energy $E = 0$; Fig. 1). Dipole transitions to the nearest metastable states with configurations $4d^9 5s\,{}^3D_3$ ($E = 6564$ cm^{-1}), $^3D_2$ (7755 cm^{-1}), $^3D_1$ (10094 cm^{-1}) and $^1D_2$ (11722 cm^{-1}) are forbidden by parity [12]. At temperatures of a vapour source up to 2000 K (resistive evaporation), the metastable states are weakly populated (over 90 % of atoms are in the $^1S_0$ state), and hence the palladium ground state $^1S_0$ should be used as the initial state in the photoionisation scheme.

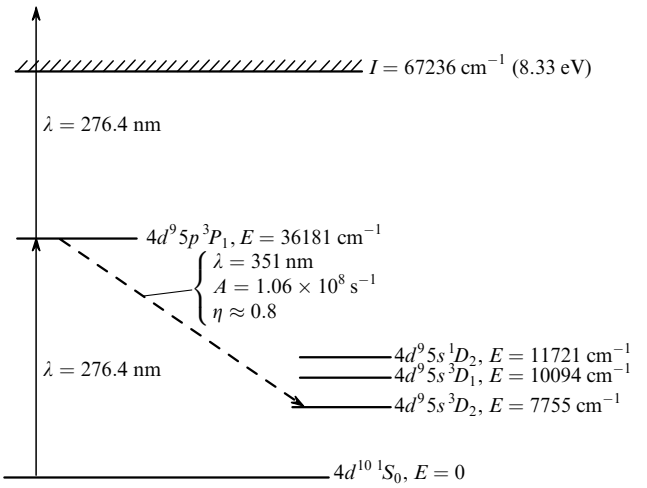

Figure 1. Pd photoionisation scheme.

The $4d^9\,5p$ configuration levels $^3P_1$ ($E = 36180.64$ cm^{-1}, $\lambda = 276.4$ nm), $^1P_1$ (40838.8 cm^{-1}, 244.8 nm), and the $^3D_1$ level (40368.73 cm^{-1}, 247.6 nm) are the levels closest to the ground state, to which dipole transitions can occur (see Ref. [13] for data on the level energies, and Ref. [14] for data on the radiative transition probabilities). A frequency-doubled dye laser pumped by a copper laser can excite only the $^3P_1$ level, which was used as the first working level upon step ionisation performed in Ref. [10] and our experiments. Unfortunately, the data presented in Refs [13, 15] indicate

that self-ionisation states are unlikely to emerge in the course of photoionisation from the $4d^9 5p\,^3P_1$ state by laser radiation at 276.4 nm.

In order to predict the required laser power for the second step, one should know the cross section of photoionisation from the excited $^3P_1$ state. For this purpose, we performed two-step ionisation of palladium by a high-power laser at 276.4 nm.

3. Description of the experiments

Fig. 2 shows the experimental setup for measuring the photoionisation cross section of palladium. It consists of a copper vapour laser (a master oscillator and three amplifiers based on Kristall LT-40CU laser tubes, a single-channel dye laser, and a vacuum chamber with an evaporator and an ion detector.

Radiation from the copper vapour laser with an output power of about 40 W at 510 nm was used for pumping the cells in the dye laser system. The pump pulse FWHM duration was 20 ns, and the laser pulse repetition rate was 10 kHz. The dye cells were pumped transversely.

Narrow-band laser radiation tunable near 552.78 nm was emitted by the dye (rhodamine 110) laser and used for excitation and photoionisation of palladium isotopes after frequency doubling. The master oscillator was based on an LZhI-504 commercial laser with an intracavity Fabry–Perot etalon with an enlarged base, which reduced the emission linewidth to 700 MHz for an average output power of 150 mW. The emission linewidth without the etalon was 15 GHz. The laser radiation was amplified to an average power of about 10 W by using a preamplifier and two amplifiers.

The wavelength of the laser radiation was measured with an accuracy of ± 0.00001 nm with a four-channel wavelength meter developed at the General Physics Institute, Russian Academy of Sciences. Absolute calibration of the wavelength meter was carried out by using the radiation from a stabilised He–Ne laser. The error of spectral measurements in the experiments did not exceed ± 0.00002 nm. The drift of the laser radiation frequency caused by a change in the temperature of the laser system components and the pressure of the surrounding medium was compensated manually by periodically adjusting the tilt of the intracavity etalon.

UV radiation at the wavelength required for excitation and photoionisation of palladium isotopes was produced by frequency doubling with the help of a nonlinear BBO crystal of length 10 mm and an aperture 4×5 mm. The laser beam was focused at the crystal by a lens of focal length 300 mm. A second lens, placed behind the crystal, compensated the second-harmonic astigmatism and collimated the UV radiation beam. The conversion efficiency was 20 %. The average UV radiation power with an approximately Gaussian intensity distribution over the beam cross section achieved 1.6 W. For a higher radiation power, the intensity distribution in the beam deteriorated noticeably.

The UV radiation beam passed through a calibrated

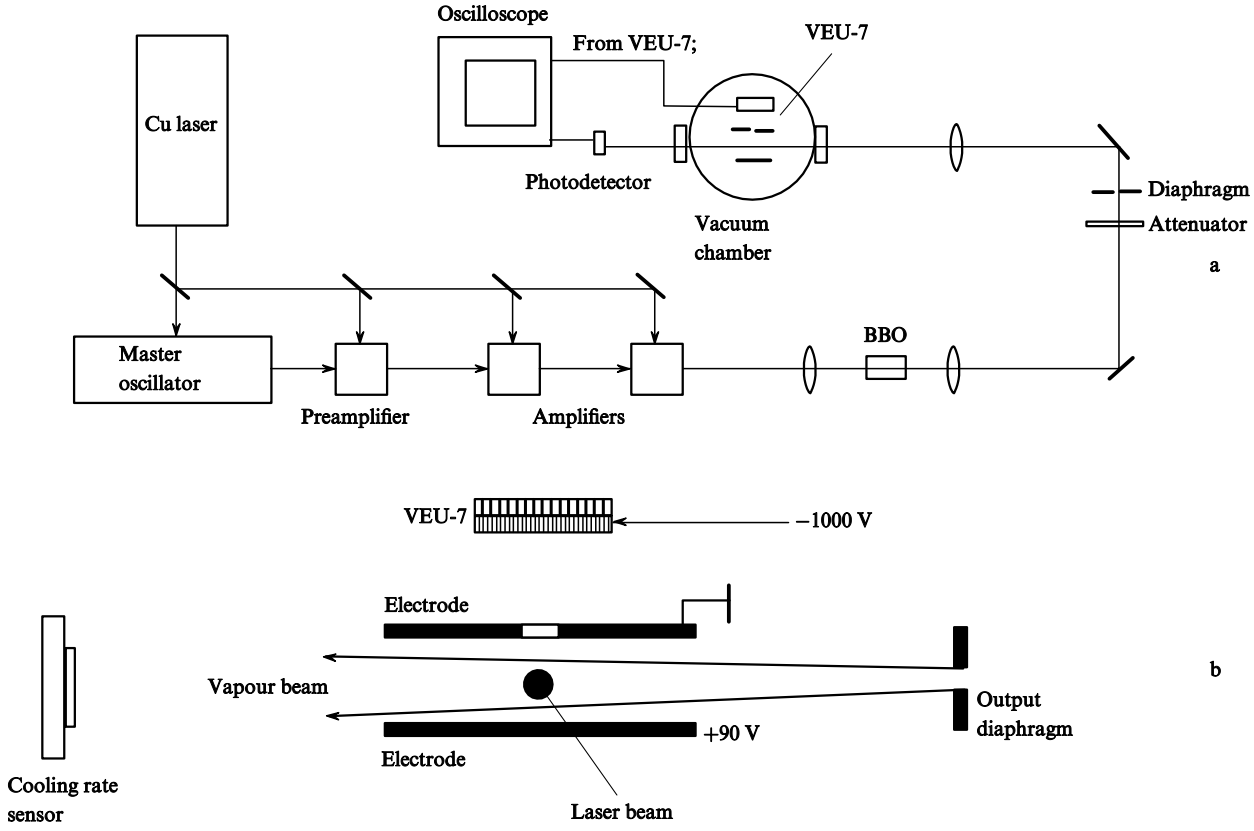

Figure 2. Schemes of the experimental setup for measuring the palladium photoionisation cross section (a) and the equipment in the measuring chamber (b).

diaphragm, which separated the central homogeneous part of the beam, and a calibrated 12-step neutral attenuator, after which it entered the vacuum chamber. The average power density of laser radiation in the region of interaction with vapour was comparatively low and did not exceed 10 W cm^{-2}. The use of a focusing lens increased the power density in the interaction region by a factor of 50 – 100, the beam diameter in this region being determined by the effective diameter of the waist at the lens focus (0.2 mm).

A resistive molybdenum evaporator was used in the experiments with palladium. The atomic beam was formed by a system of diaphragms. The angular divergence of the atomic beam did not exceed 0.1 rad, which made it possible to reduce the Doppler width of the atomic absorption lines of palladium down to 300 MHz (compared to the width 2.77 GHz in the uncollimated vapour of palladium atoms). The density of atoms in the interaction region was about 10^9 cm^{-3}. The vapour density was estimated from the rate of deposition of palladium atoms on a quartz crystal (a INFICON sensor of the rate of film deposition) placed in the palladium vapour beam at a distance of 55 mm from the interaction region. The chamber was evacuated by oil-free pumping down to 10^{-5} Torr.

The second-harmonic emission spectrum covered the isotopic structure of palladium, so that photoionisation was nonselective, and ions of all isotopes of palladium appeared in the interaction region. The ions emerging in the interaction region during the action of the laser radiation pulse were extracted from the plasma by an electric field over a period smaller than the laser pulse repetition period and directed at a VEU-7 electron multiplier through a hole in the electrode. The ion current signal from the electron multiplier was displayed with a Tektronix TDS 3034 digital oscilloscope. The average UV radiation power was measured at the chamber entrance. Fig. 3 shows a typical ion current saturation curve in our experiments.

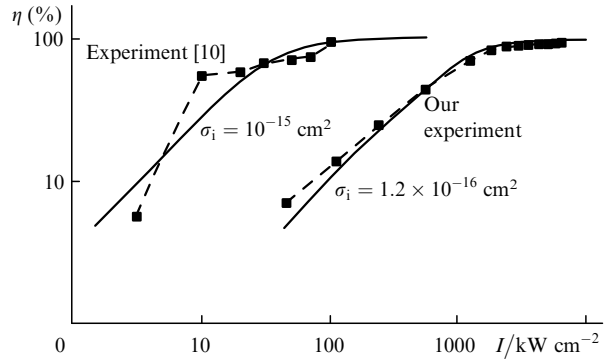

Figure 3. Theoretical dependences of the ionisation efficiency on the laser pulse intensity (solid curves); experimental points correspond to values of the photocurrent normalised to the theoretical curves corresponding to the maximum experimental intensities.

4. Calculation of the two-step ionisation efficiency

If the $^3P_1$ state is excited and ionised by a single laser with a homogeneous intensity distribution over the beam cross section, the number of atoms ionised per pulse can be determined from the rate equations:

$$\frac{dN_0}{dt} = N_1[A + B_0 f(t)] - N_0 B_0 f(t),$$

$$\frac{dN_1}{dt} = -N_1[A + \gamma + (B_0 + B_1) f(t)] + N_0 B_0 f(t), \qquad (1)$$

$$\frac{dN_i}{dt} = N_1 B_1 f(t).$$

Here, N_0 and N_1 are the populations of the ground (0) and excited (1) states; N_i is the density of ions; A is the rate of the $1 \to 0$ spontaneous transition; γ is the total rate of transition of atoms from the excited state to lower metastable states, which are neglected in the model; $B_0 = I_0\sigma_0/\hbar\omega$ is the rate of laser-induced $1 \to 0$ transition; $B_1 = I_0\sigma_i/\hbar\omega$ is the rate of ionisation of the excited state by the laser radiation; I_0 is the average intensity of a laser pulse; σ_0, and σ_i are the photoexcitation and photoionisation cross sections, respectively; $f(t)$ is a function describing the time dependence of the laser pulse; and $\hbar\omega$ is the energy of a laser photon.

We used the time dependence of the laser pulse in the form

$$f(t) = \frac{1}{\sqrt{\pi}} \exp\left[-\left(\frac{t - 2\tau}{\tau}\right)^2\right].$$

The values of the parameters were: $\tau = 10$ ns, $\hbar\omega = 4.5$ eV, $A = 1.03 \times 10^7$ s^{-1}, $\gamma = 1.28 \times 10^8$ s^{-1}, and $\sigma_0 = 2 \times 10^{-13}$ cm^2. The value of $I_0\sigma_i$ was varied. It was assumed that all the atoms are in the ground state at the initial instant $t = 0$. The ionisation efficiency $\eta = N_i(\infty)/N_0(0)$ was determined by solving the system of differential equations (1).

The calculated and experimental results are presented in Fig. 3. One can see that the experimental results described in Ref. [10] can be interpreted theoretically by assuming that the photoionisation cross section is very large ($\sigma_i \sim 10^{-15}$ cm^2). Such values are not characteristic of the photoionisation cross sections. Near the ionisation threshold, photoionisation cross sections are usually of the order of 10^{-17} cm^2. We are unable to explain why the ionisation efficiency attains saturation for such a low radiation intensity ($I_0 \approx 10 - 40$ kW cm^{-2}) in Ref. [10]. The results of our experiments are interpreted quite well by the theory for $\sigma_i = 1.2 \times 10^{-16}$ cm^2. This value appears to be more reasonable.

5. Conclusions

We have shown that to obtain the efficient (more than 50 %) ionisation of the $4d^9 5p\,^3P_1$ state of palladium by 276.4-nm, 10-ns laser pulses with a pulse repetition rate of 10 kHz, the average radiation power should exceed 200 W cm^{-2}. Calculations show that a comparatively moderate intensity of 0.1 W cm^{-2} is sufficient for the saturation of the first $4d^{10}\,^1S_0 - 4d^9 5p\,^3P_1$ transition. The question of the isotopic selectivity of the two-step ionisation scheme of palladium used by us requires independent investigation.

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