Laser photochemical lead isotopes separation for harmless nuclear power engineering

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Abstract. The collisional quenching of the metastable $^3P_{1,2}$ and $^1D_2$ lead atoms is studied experimentally in the gas flow of the lead atoms, reagent-molecules and a carrier gas Ar. The experimental parameters were similar to the conditions that are required in the operation of the experimental setup for photochemical isotope separation. Excited atoms are generated under electron impact conditions created by a gas glow discharge through the mixture of gases and monitored photoelectrically by attenuation of atomic resonance radiation from hollow cathode $^{208}$Pb lamp. The decay of the excited atoms has been studied in the presence various molecules and total cross section data are reported. The flow tube measurements has allowed to separate the physical and chemical quenching channels and measure the rates of the chemical reaction excited lead with $N_2O$, $CH_2Cl_2$, $SF_6$ and $CuBr$ molecules. These results are discussed in the prospects of the obtaining isotopically modified lead as a promising coolant in the reactors on the fast-neutron.

1. Introduction
The great interest currently exists to isotopically modified lead as a promising coolant in fast-neutron nuclear power plants. It is assumed that the heavy metal coolant and the proton-to-neutron converter will be used (lead or lead–bismuth alloy). However, long-term irradiation of lead formed an extremely dangerous $\alpha$- and $\gamma$- radionuclides, including $^{210}$Po and $^{207}$Bi. It was shown that a coolant enriched by isotope $^{208}$Pb (up to the abundance of 99.0%) produces far less (by a factor of $10^3 \div 10^4$) polonium and bismuth radionuclides than natural lead [1].

The technologies to produce the superpure and isotopically modified lead are well developed. Unfortunately, the cost of lead isotopes depending on the production method is not suitable for modern power engineering. The necessity for isotopically modified lead can be thousands tons already within 10 years. Therefore, the development of alternative technologies leading to the production of a considerably (by orders) less cost product is actual. The laser isotope separation has the significant advantages [2, 3] compared with other known methods. They are as follows a high selectivity of an elementary separation act, the low energy expenditures, the possibility of the target isotope isolation, non-contact of the separation method. When two types of atoms of different isotopic composition ($^1A$ and $^2A$) have even one spectral line that does not overlap with others, it is possible to excite selectively with laser light an atom or molecule ("particle" $^1A$) of a needed isotopic composition. The excitation of the $^1A$ particles changes their chemical and physical properties and, hence, may be used to separate substances by any method based on differences in the characteristics of excited and unexcited...
particles. Selective laser excitation of \(^{1}\)A particles is followed by the chemical reaction of \(^{1}\)A* to give products designated \(^{1}\)AR. The chemical reactivity of atoms or molecules is often increased by excitation. For laser isotope separation, long-living (metastable) states are preferred. Indeed, the chemical interaction probability for an excited atom should exceed the probability of radiation decay of the level, that is, the condition \(k' n > \tau_0^{-1}\) must be satisfied. Where \(k'\) is the rate constant of the reaction, \(n\) is the concentration of reagent molecules and \(\tau_0\) is the radiation lifetime. Laser can be used to prepare reagents in particular quantum states. The excitation of a reagent can have several effects on a chemical reaction. Laser excitation of a reactant atom not only increases its internal energy, it also generally modifies its electronic state symmetry. It is well known that symmetry plays an important role in photon-induced but it can also play an important role in chemical reactivity. Electronic excitation invariably changes the shape and symmetry of the potential energy surface (PES) and it may induce a different reaction mechanism compared with that of the ground state. Possibility of the photochemical method of lead isotope extraction is based also on the peculiarities of a low electron level structure of lead. He has metastable states with the life time of \(10^{-1} \div 10^{-2}\) s, which suits ideal for conducting the photochemical reactions. The second peculiarity is that the transitions inside the configuration are hit into radiation of the highly effective semiconductor and other lasers. It gives the perspective to simplify and to make cheaper the technology of lead isotope production.

Collisions of metastable lead atoms with various molecules investigated in this work. The use of the flow tube measurements [4-6] has allowed us to separate the physical and chemical quenching channels and measure the rates of these quenching processes. The scheme of these experiments is similar to the real system for laser isotope separation. A considerable amount of data on the quenching of he optically metastable states \(^{131}\)Pb\((6\,^1D_2)\), \(^{131}\)Pb\((6\,^1S_0)\), \(^{133}\)Pb\((6\,^3P_1)\) and \(^{133}\)Pb\((6\,^3P_2)\) has already been reported [7-10]. Total thermal deactivation rate constants, but not product channels, for all excited states have been measured with some molecules at room temperature. The main objective of this study is to justify a physical possibility of using the photochemical laser method for lead isotope separation. For this purpose some experimental work should be carried out to find the effective chemical reactions of the metastable \(Pb\) atoms with molecules.

### 2. Description of the method

The gas flow apparatus for the laser isotope separation is shown in Figure 1. Collisional reactions of the metastable lead atoms and reagent take place in the interaction chamber constructed of cylindrical tube with inner cross section area \(S\) and long \(L\) having two end windows.

![Figure 1. Experimental setup for photochemical isotope separation of lead atoms.](image)
The section of the reactor on the reaction zone was heated by an external furnace to prevent the deposition of lead on tube walls. The lead atoms in a mixture with a carrier gas were introduced into the region of interaction from a heated reservoir. By varying the reservoir temperature, we could vary the concentration of lead atoms in the interaction zone. The carrier gas (Ar) is the major component in the flow tube and thus serves to define the physical properties of the gas flow. The reagent-molecules are mixed homogeneously with the carrier gas. The gas mixture was evacuated at the pumping rate G by a vacuum pump. The gas flow velocity is \( v = G \cdot S^{-1} \). The propagation time of the lead atoms through the interaction area is \( T = L \cdot v^{-1} \). To excite the lead atoms the monochromatic laser radiation propagates along the gas flow. Long-lived (metastable) states produced via the forbidden transitions. They can lose their excitation energy by collisions with molecules and other particles by the action of various physical and chemical processes in the interaction chamber. These processes we have combined only in three channels:

\[
\begin{align*}
  Pb^* & \rightarrow Pb + h\nu \quad Pb^* + \text{(wall)} \rightarrow Pb \\
  Pb^* + XY & \rightarrow Pb + (XY)^* \\
  Pb^* + XY & \rightarrow PbX + Y
\end{align*}
\]  

Channel (1a) contains contributions to the lifetime \( \tau_0 \) of metastable atoms in the absence of a reagent. Channel (1b) is the transfer of excitation energy to the internal and translational degrees of freedom of the colliding partner proceeding at a rate constant \( k_{ip} \) (physical quenching). Lead atoms in the ground state are formed by such processes. Channel (1c) is the collisions of the metastable atom \( Pb^* \) with XY molecules which leading to a chemical reaction. Lead atoms combine with molecule into stable chemical compounds with the value the rate constant \( k_{ic} \) (chemical quenching) and lifetime is more than \( T \). The chemical compounds of \( Pb \) atoms with other particles have other chemical and physical properties. The stable reaction components are deposited on the walls of the interaction chamber or move along with the flow. In such a way, this process is characterized as depletion atoms in the reaction zone. From the interaction chamber the gas flow arrives in the collector chamber. By selecting the walls temperature can be done there the deposition of the reaction products. The time interaction of the \( Pb \) atoms with the laser radiation can be changed by varying the pumping rate. The flow tube technique is a good method to measure the kinetic parameters of the reactions. It allows determining the individual values for the rate constants of the physical and chemical quenching. The solution of the rate equation for the concentration of the lead atoms \( N(T) \) in the interaction chamber is presented by the expression:

\[
N(T) = N_0 \cdot e^{-T[k_{ip} + \Sigma(W_i \cdot \theta_i)]},
\]  

where \( T \) is the propagation time of the lead atoms through the interaction area, \( N_0 \) is the concentration of the lead atoms in the interaction chamber with \( T = 0 \), where the atoms and gas-reagent are mixed, \( n \) – concentration of reagent-molecule, \( k_0 \) - rate constant of chemical reaction between ground state lead atoms and reagent. The rate of atomic excitation \( W_i \) of the \( i \) metastable state per second under action laser radiation or other processes, the decay rate \( R_i \) of \( i \) metastable atoms in the interacting volume and the efficiency (probability) \( \theta_i \) of the formation chemically stable product in process (1c) is given by the relations:

\[
W_i = R_i \left( \frac{N_i(T)}{N(T)} \right), \quad R_i = \left( \frac{1}{\tau_{0i}} + k_i \cdot n \right), \quad \theta_i = \frac{k_{ic} \cdot n \cdot \tau_{0i}}{1 + (k_{ip} + k_{ic}) \cdot n \cdot \tau_{0i}}
\]  

where \( N_i(T) \) is the stationary concentration of \( i \) metastable lead atoms in the interaction chamber, \( \tau_{0i} \) is the lifetime of \( i \) metastable atoms in the absence of a reagent, \( k_i = k_{ic} + k_{ip} \) is the total second-order rate constants for chemical reaction and physical quenching.
3. Selective excitation of the metastable lead atoms

The possible optical schemes and properties of a long lived level excitation are presented in Figure 2 and Table 1. The 6s6p5 ground configuration of Pb consists of five levels: 1P0.1, 1D2, and 1S0. Since electric-dipole (E1) transitions between states of the same parity are forbidden, all levels of these configurations are metastable. In the second-order perturbation theory weak magnetic-dipole (M1), electric-quadrupole (E2), or mixed (M1+E2) type transitions between these levels are allowed. Lead occurs in nature in the form of four different stable isotopes of masses 204, 206, 207, and 208. For 207Pb the nuclear spin quantum number is I=1/2, and only this isotope shows a hyperfine structure. The isotope shifts of multipole lines are relatively small in comparison with resonance on (λ = 283.3 nm) [11-13]. All known values are presented in Table 1. Figure 2 shows the possible excitation schemes of the metastable lead atoms by one and two step way. The rate atomic CW excitation of the atom Wt in the flow is determined by the equation:

\[ W_t = \frac{\sigma \cdot P}{S_L \cdot h \cdot v_0}, \]

where P is power, S_L is cross-sectional area and v_0 is central frequency of radiation transition. The excitation cross sections for the metastable states are small and have values \( \sigma \leq 10^{-17} \div 10^{-19} \text{ cm}^2 \).

| Transition   | λ, nm | \(^a\lambda_i\), s\(^{-1}\) | \(^b\sigma\), cm\(^2\) | \(^c\Delta\nu_i\), MHz | \(^d\Delta\nu_a\), MHz | \(^d\Delta\nu_a\), MHz | \(^d\Delta\nu_a\), MHz |
|--------------|------|----------------------------|-----------------|-------------------|------------------|-------------------|------------------|
| \(^3\)P\(_0\)\rightarrow\(^7\)s\(^1\)P\(_1\) | 283.3 | 4.7×10\(^{-7}\) | 2.6×10\(^{-12}\) | 1600 | 10277.9 | 2317.6 | 4292.5 | 2922.1 |
| \(^3\)P\(_0\)\rightarrow\(^3\)P\(_1\) | 1279.3 | 7.9 | 4.1×10\(^{-17}\) | 352 | -1066.7 | -220.4 | - | 2533.3 |
| \(^3\)P\(_0\)\rightarrow\(^1\)P\(_2\) | 939.2 | 0.49 | 1.7×10\(^{-18}\) | 478 | - | - | - | - |
| \(^3\)P\(_0\)\rightarrow\(^1\)D\(_2\) | 446.2 | 0.17 | 7.3×10\(^{-20}\) | 957 | - | - | - | - |
| \(^3\)P\(_2\)\rightarrow\(^1\)S\(_0\) | 531.6 | 18.7 | 4.6×10\(^{-19}\) | 846 | - | - | - | - |
| \(^3\)P\(_1\)\rightarrow\(^1\)D\(_2\) | 733.4 | 14.5 | 8.1×10\(^{-18}\) | 614 | -1320 | -184 | 334 | 2117 |
| \(^3\)P\(_2\)\rightarrow\(^1\)D\(_2\) | 925.6 | 10.6 | 7.3×10\(^{-18}\) | 480 | - | - | - | - |

\(^a\) probabilities of spontaneous emission from level \( \lambda \) to \( i \).
\(^b\) cross-section at center of the Doppler absorption contour.
\(^c\) Doppler width of the absorption contour at 930K°.
\(^d\) shift between two isotopes of Pb.

This circumstance necessitates the exciting at high laser power, the major part of which may be lost due to a relatively low absorption. A multipass system is used as a technical solution. Very attractive way of selective production the metastable excited state if the lead atoms were excited from the ground state 6p\(^3\)P\(_0\) to the 6p7s\(^1\)P\(_1\) resonance state under action monochromatic laser radiation at (λ= 283.3 nm) with lifetimes 5.8 ns. Then 6p7s\(^1\)P\(_1\) resonance state decay into the ground and well into the metastable levels by the superluminescence. In this case the laser part of the experimental setup is more expensive than the other methods. For isotope separation by photochemical method the isotope atom concentration is restricted by the process of resonance energy transfer from an atom of one isotope to an atom of another one. This process obviously acts as the excitation selectivity deterioration. Excitation transfer cross-section in the approximation of the dipole-dipole interaction is in the inverse proportion to the life time of the upper level. The long-lived states have the small values of the dipole moment and a cross-section excitation \( \sigma \leq 10^{-17} \text{ cm}^2 \) (Table 1). The rate of energy transfer to another isotope (\( \sigma \nu_\lambda N \)) must be less than (1/T). Here \( \nu_\lambda \) is the arithmetic mean velocity of
the lead atoms. This gives the upper limit of the atomic concentration $\sim 10^{13}$ cm$^{-3}$. This value is by three orders of magnitude higher than the limiting atom concentration admitted in photoionization isotope separation method. The main peculiarity is that the transitions between metastable states are hit into radiation of the highly effective semiconductor and other lasers. It gives the perspective to simplify and to make cheaper the technology of lead isotope production. Laser isotope separation of lead atoms in the gas flow is effective even if the corresponding isotope structure is masked by the Doppler broadening. In this case, the selectivity of excitation increases at frequency detuning from the lines of all isotopes. This demonstrates Figure 3, which shows the total and Doppler absorption contour of each Pb isotope. The narrowband laser radiation can be used to provide velocity selective excitation of the inhomogeneously broadened absorption contours. For a mixture of isotopes, the absorption contour represents the sum of values with corresponding $\nu_0$ and weights proportional to the isotope percentages. If the laser frequency is tuned to the isotope absorption edge, the contribution from other isotopes becomes small. In this case, decreases sharply the total absorption coefficient, but the large interaction time $T$ between the laser radiation and atoms provides 100% excitation efficiency for the desired isotope.

**Figure 2.** Energy levels of atomic lead revelant to this paper. All wavelength transition are given in nm.

**Figure 3.** Lead atom absorption spectrum at the transition $6p^2\, ^3P_0-6p^2\, ^3P_1$ ($\lambda = 1279.3$ nm).

4. Experiment and results
The wall temperature of the interaction chamber was 930° K and argon concentration is 4·10$^{16}$ cm$^{-3}$. Gas mixture of the molecules, carrier gas and Pb atoms was evacuated at a rate $G = 0.6$ l·s$^{-1}$ by a pump with velocity is $v = 90$ cm·s$^{-1}$. Lead atoms were excited under electron impact conditions created by DC or pulsed glow discharges through the mixture of gases. The length of the discharge gap is 10 cm. The reaction time $T = 0.11$ s. The concentration of the Pb atoms was determined by absorption measurements of the transmitted intensity of the resonant radiation from $^{208}$Pb lamp through the interaction region obeyed the Beer’s law. For the ground state measurements were used the $6p^2\, ^3P_0 \rightarrow 7s\, ^3P_1^0$ ($\lambda = 283.3$ nm) resonance transition. For measurements the populations of the excited states were used other spectral lines. The output radiation from $^{208}$Pb lamp focused into the entrance slit of a
monochromator. He was used as a band-pass filter and tuned to some emission lines of Pb. Measurements were carried out in two cases transverse direction to the gas flow. The first direction is located in the discharge region. The second direction is located beyond the region of the interaction of excited atoms and molecules. The light-induced fluorescence (LIF) method is used for the temporal behavior population of metastable lead atoms in the interaction chamber. Under action of resonance radiations the Pb atoms occur the optical transitions from the metastable states to the $7s\ ^3P_1^0$ level (Figure 1). The time dependent fluorescence $S_i(t)$ at 405.78 nm ($7s\ ^3P_1^0 \rightarrow 6p\ ^2P_2$) was recorded from illuminated area. The multiplier signal $S_i(t)$ is proportional intensity of this fluorescence and, accordingly, metastable state population determined by the equation:

$$S_i(t) = C \cdot e^{-(R_i t)},$$

where $C$ is a time independent constant. The study of the metastable state quenching in collisions with molecules were carried out using the afterglow pulsed discharge with a repetition rate of 100 Hz and current pulse with half-width of 4 µs. The output multiplier signal $S_i(t)$ processed by a box-car integrator and stored in a computer at various molecules concentrations and delay times $t$ between the resonance radiations and the discharge pulse. From the decay curves (5) were determined the values of $k_0$ and $k_i$. For $^1D_2$ metastable levels the decay curves of $S_i(t)$ were measured by the interaction of the Pb atoms with pulsed Pb vapor laser radiation (722.9 nm) with half-width of 10 ns and pulse energy of 10 µJ. For monitoring $6p\ ^23P_1$ and $6p\ ^23P_2$ metastable levels was used two sharp spontaneous emission lines emitted from the active medium of the Pb vapor laser at 363.96 and 405.78 nm, belonging to the $7sP_1^0 \rightarrow 6p\ ^23P_1$ and $7sP_1^0 \rightarrow 6p\ ^23P_2$ transitions, respectively. The light pulses were synchronized in time with discharge pulses. Plots for the obtained quenching cross sections for Pb($^3P_1, \ ^3P_2, \ ^1D_2$) atoms versus the serial number of interacting molecules are shown in Figure 4. This figure does not reveal the correlated behavior of the quenching cross sections for different metastable. The data for $6^3P_2$ state can be divided into three groups on the basis of the magnitude of the quenching cross section. Weak quenchers, including $H_2O$, $NH_3$, and $C_2H_5OH$ all have quenching cross sections less than 1 Å². Strong quenchers, including $(C_2H_5)_2O$, $CH_2Cl_2$ and $CuBr$ have quenching cross sections in excess of 10 Å². Whereas the rest of the molecules have quenching cross sections values between 1 and 10 Å². This division might indicate in a general way that several different mechanisms must be considered for the quenching process.

![Figure 4. Quenching cross sections for excited Pb($^3P_1, \ ^3P_2, \ ^1D_2$) by various molecules collisions at 930 K. Serial number of interacting molecules: 1 - Water ($H_2O$). 2 - Ammonia ($NH_3$). 3 - Ethanol ($C_2H_5OH$). 4 - Propane ($C_3H_8$). 5 - Isobutanol ($C_4H_{10}O$). 6 - Benzene ($C_6H_6$). 7 - Carbon dioxide ($CO_2$). 8 - Methanol ($CH_3OH$). 9 - Ethyl bromide ($C_2H_5Br$). 10 - Nitrous oxide ($N_2O$). 11 - Acetone ($CH_3COCH_3$). 12 - Sulfur hexafluoride ($SF_6$). 13 - Diethyl ether ($C_4H_{9}O$). 14 - Methylene chloride ($CH_2Cl_2$). 15 - Copper bromide ($CuBr$).]
Chemical contributions to the total collisional quenching are investigated by the reduction of lead atom concentration due to chemical reaction for excited atoms with molecules. Relative atomic concentration measurements were carried in the second direction of the flow during the presence and absence of a glow discharge in the interacting region. In these measurements, the metastable lead atoms were excited in a CW glow discharge with current 50 mA. Simultaneously with photochemical reaction (1c) occur chemical reaction between of lead atoms in the ground state and added molecules with the rate constant \( k_0 \). For some molecules the reaction rates of these two processes can be compared with each other. Nearly all chemical reactions have the activation barrier. Table 2 lists the results of our measurements for the values which describe the depletion excited lead atoms in the interaction chamber by chemical reaction. Unexpectedly, but only for four of the fifteen investigated molecules the change in concentration of the Pb atoms was revealed.

| Molecule | \( k_0^{ab}, 10^{13} \) | \( \sum (W_i, \theta_i), s^{-1} \) | \( k_w/k_{ip} \) | \( k_w, 10^{12} \) | Product | \( \Delta H^o, eV \) |
|----------|-----------------|-----------------|-----------------|-----------------|---------|-----------------|
| \( N_2O \) | 0.01 | 14.3, [2·10^{-3}]c | 0.28 | 0.93 | PbO | -2.31 |
| \( CH_2Cl_2 \) | 8.2 | 5.2, [10^{-3}]c | 0.043 | 24 | PbCl | 0.38 |
| \( SF_6 \) | 0.0016 | 4.1, [6.8·10^{-3}]c | 0.031 | 1.3 | PbF | 0.27 |
| \( CuBr \) | 4.1 | 3.3, [4.5·10^{-3}]c | 0.025 | 70 | PbBr | 0.83 |

- Second-order rate constants, units \( cm^3·s^{-1} \).
- Reference 6.
- Pressure gas-reagent [Torr].

For eleven other molecules studied, the effect of depletion did not exceed measurement errors in spite of the high rate constants \( k_i \) for quenching. This means that the collisions of metastable lead atoms with these molecules caused effective quenching of electronic excitation energy without noticeable chemical reactions (\( k_c \ll k_{ip} \)). The extent of chemical quenching is reflected in the depletion in the ground state lead atom concentration measured sufficiently long after collisions to allow for complete relaxation of all excited species. Our gas flow apparatus is allows studying chemical reactions with half-lives of \( T \approx 0.11 \) sec or longer. This time is longer than the time of the radial diffusion \( 4·10^{-2} \) s for excited species on the walls of the interaction chamber where they effectively relax to the ground state before reaching the observation region. Very many lead atomic states are excited in the gas discharge, which participate in the collision process. To find the individual contribution of the each excited state, we consider the required conditions for efficient chemical reaction. According to equation (2) high efficiency depletion of the \( i \) lead atoms states is determined by the condition \( (\theta_i·W_i) \geq 1 \). From the experimental data we can determine only total sum of values \( (\theta·W) \) for all possible states. The short-lived and high-lying states give a minor contribution to the depletion of lead atoms, because the condition \( (k_i · n_0) \leq 0.1 \) is always satisfied for them. This condition requires another condition \( (\theta \ll 1) \), which follows from the expression (3). From the relation (2) follows, that under this condition the efficiency of the chemical reaction is low. The metastable Pb atom states produced in the discharge are mostly a combination of four \( ^3P_1, ^3P_2, ^1D_2 \) and \( ^3S_0 \) states. All considered chemical reactions are exothermic. It is believed that only the lowest metastable states are formed in significant amount, since all higher lying states will strongly radiate to these states. The minor contribution of the \( ^1D_2 \) and \( ^3S_0 \) states in a chemical reaction is related with exclusions by the Wigner rules [14]. Thus, only two metastable states \( ^3P_1 \) and \( ^3P_2 \) produced in the glow discharge can effectively react with molecules to yield stable products. They are effectively excited in a gas discharge (their stationary concentration amounts to 20% of the concentration of the ground.
state) and possess long lifetimes with respect to spontaneous decay. Between these two states should choose only $^3P_2$ state, for which the activation barrier is smaller and hence the chemical reaction is more efficient. Table 2 lists the values for rate constants for $k_e$ and $k_q$ at $W_i = 144$ s$^{-1}$, $\tau_0 = 10^{-3}$ s. More accurate data can give the experiments with selective excitation. The choice of molecules as reagent in the present experimental study was motivated by the low value of $k_q$ and by the ecological purity of those substances. These properties are necessary for laser isotope separation.

5. Discussion

The experimental results allowed us to forecast prospects of the photochemical lead atoms isotope separation. The appropriate power continuous wave laser radiation is $\lambda = 1.3$ µm, $P = 10$ W and a low line width of $\leq 10$ MHz is use. The number of photons arriving per second is $K \approx 6 \times 10^{19}$ photon/s. This yields the rate of isotope separation $H = 650$ kg/year for the 100% quantum efficiency of the separation process (from one photon comes one atom of the desired isotope). In reality, this value is much less. It is defined the real physical and chemical processes that take place in the interaction chamber. We adopt the following parameters for the interaction chamber: the cross section area and length of the interaction chamber are $S = 10$ cm$^2$, $L = 10^2$ cm respectively, the drift velocity of the particles in the gas flow is $v = 10^5$ cm s$^{-1}$. Then the interaction time of lead atoms with laser radiation is $T = 1$ s. The following concentrations values are acceptable in the gas flow: $N = 10^{13}$ cm$^{-3}$, $n = 10^{13} \div 10^{15}$ cm$^3$, $5 \times 10^{16}$ cm$^{-3}$ for the lead atoms, reagent-molecules and argon atoms respectively. The number of atoms per second in the gas flow moving through the cross-section $S$ is equal to $F = NvS = 10^{16}$ atoms s$^{-1}$. At 100% excitation of lead atoms we obtain the rate of isotope separation $H = 0.1$ kg year$^{-1}$. Let us consider excitation of lead atoms at the optical transition $6p^2\Sigma^3P_0 \rightarrow 6p^2\Pi^3P_1$ ($\lambda = 1.279$ µm). From relation (4) we find the rate of atomic excitation under action resonance laser radiation $W_i = 2.5 \times 10^2$ s$^{-1}$ at the center of the absorption line with $\sigma = 4.1 \times 10^{17}$ cm$^2$ (Table 1). This implies that during the time of the movement of the atoms into the interaction region of the laser excitation probability is $W_iT = 2.5 \times 10^2 >> 1$. That is, the atom can be excited multiple times in the interaction chamber. Even for the detuning laser frequency at the edge of the absorption line (Figure 3), the probabilities of atomic excitation remain a large enough. The quantity $\theta_i$ in the relation (2) is defined as the ratio of the decay rate of chemical quenching of metastable atoms to the overall quenching decay rate. This quantity can assume values in the range $0 \leq \theta \leq 1$. The results of the Table 2 show that the most effective chemical reaction is realized with $N_2O$ molecule, for which $\theta = 0.1$. At 100% efficiency of the laser excitation for the metastable lead atoms in the flow and the same efficiency of decay rate. This quantity can assume values in the range $0 \leq \theta \leq 1$. The results of the Table 2 show that the most effective chemical reaction is realized with $N_2O$ molecule, for which $\theta = 0.1$. At 100% efficiency of the laser excitation for the metastable lead atoms in the flow and the same efficiency of the chemical reaction can be realized the production rate of the necessary isotopes. The obtained results allow one to define the range of the possible chemical reagents for laser isotope separation of lead by photochemical method. The $6p^2\Sigma^3P_2$ lead state can be excited isotopically selectively in $6p^2\Sigma^3P_0 \rightarrow 6p^2\Sigma^3P_2$ scheme by radiations from semiconductor laser, operating at 939 nm. Gas flow conditions allow multiple radiation interactions with atoms to be performed with high efficiency in spite of small cross-section transition and low laser power. The obtained results allow one to define the range of the possible chemical reagents for laser isotope separation of lead by photochemical method.

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