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POSSIBILITY OF SEPARATING THE ISOTOPES  $^{127}\text{I}$  AND  $^{129}\text{I}$  WITH THE AID  
OF A PHOTODISSOCIATION IODINE LASER

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A method for separating iodine isotopes is proposed, based on the large difference between the rate constants of the excited  $\text{I}^*(^2\text{P}_{1/2})$  and unexcited iodine atoms with radicals  $\text{CF}_3$  and with  $\text{Cl}_2$  molecules, and on the possibility of selectively acting on the  $^{127}\text{I}$  atoms in the states  $^2\text{P}_{1/2}$  and  $^2\text{P}_{3/2}$  by radiation from a photodissociation  $\text{R}^{127}\text{I}$  iodine laser ( $\lambda = 1.315 \mu\text{m}$ ). The possibility of separating the pure isotope  $^{129}\text{I}$  and the mixture  $^{127}\text{I}$  with  $^{129}\text{I}$  is investigated.

1. METHOD OF IODINE-ISOTOPE SEPARATION AND ESTIMATE  
OF THE LIMITING ENRICHMENT PARAMETERS†

An appreciable part of the presently discussed methods of laser isotope separation (LIS) is based on selective photoexcitation of the isotopic atoms or molecules, followed by binding the excited particle with a specially added reagent in a chemical reaction [1, 2]. In the general case, the choice of the chemical reaction for binding electron-excited atoms or molecules entails great difficulties, but for iodine atoms it was found to be possible to select such reactions. Moreover, the proposed isotope-separation method can apparently also be used for other atoms, since it is based principally on the difference between the rates of recombination of the unexcited and excited atoms into molecules. The difference between the recombination rates of the excited and unexcited atoms, which we have experimentally observed for iodine atoms [3], is expected to have a more general character.

In this paper, we consider an iodine isotope separation method in which it is possible to achieve an enrichment coefficient  $\alpha \geq 2$  per cycle and to obtain the required iodine isotopes with a content more than 99%.

†The main premises of this method were reported at the International Conference on Lasers and Their Applications (Dresden, 1977).

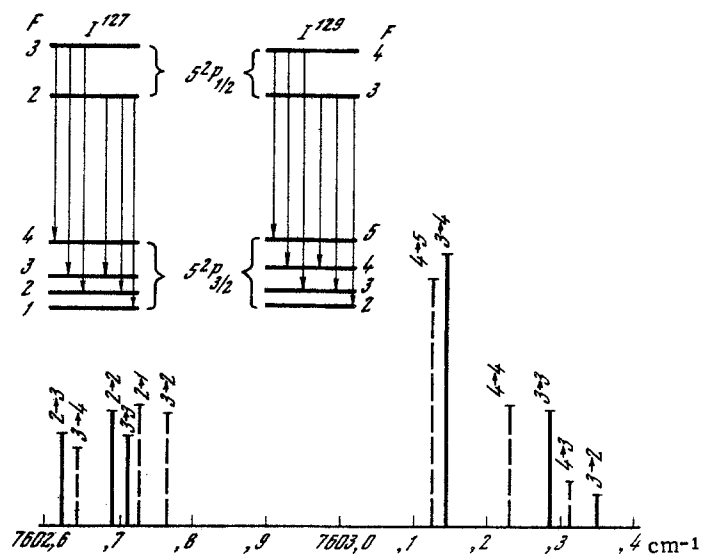


Fig. 1. Level systems and luminescence spectra of  $^{127}\text{I}$  (solid lines) and  $^{129}\text{I}$  (dashed) (the heights of the segments corresponding to the individual components are proportional to their intensities).

The output of a small laboratory setup can amount to  $10^{-3}$ - $10^{-4}$  g/h of the enriched isotope.

The method is based on the large difference between the rate constants of the reactions of the excited  $\text{I}^*(^2\text{P}_{1/2})$  and unexcited  $\text{I}(^2\text{P}_{3/2})$  iodine atoms with the radicals  $\text{CF}_3$  and the molecules  $\text{Cl}_2$ , and on the possibility of selectively acting on the iodine atoms  $^{127}\text{I}$  in the states  $^2\text{P}_{1/2}$  and  $^2\text{P}_{3/2}$  by radiation of a photodissociation iodine laser (PDIL) operating on the molecules  $\text{R}^{127}\text{I}$  ( $\lambda = 1.315 \mu\text{m}$ ).

The proposed method can be of particularly practical importance for the obtaining of pure  $^{129}\text{I}$  (half-life  $1.72 \cdot 10^7$  years [4]), inasmuch as the commercially obtainable product contains not less than 25%  $^{127}\text{I}$  [5]. Therefore, we shall hereafter only discuss the concrete method of separating the isotopes  $^{129}\text{I}$  and  $^{127}\text{I}$ .

The gist of the proposed method is the following: The initial gas mixture in the working quartz cell consists of  $\text{CF}_3^{127,129}\text{I}$ ,  $\text{Cl}_2$  and  $(\text{CF}_3\text{CO})_2\text{O}$ . The light from the pump flash lamps produces photodissociation of  $(\text{CF}_3\text{CO})_2\text{O}$  and  $\text{CF}_3\text{I}$  in the cell (to prevent photodissociation of  $\text{Cl}_2$ , the cell is surrounded by a  $\text{Cl}_2$  filter). The results are the excited atoms  $^{127}\text{I}^*(^2\text{P}_{1/2})$  and  $^{129}\text{I}^*(^2\text{P}_{1/2})$ , and the radicals  $\text{CF}_3$ . At the same time, radiation from a PDIL based on  $\text{R}^{127}\text{I}$  molecules passes through the gas. In the laser field, the  $^{127}\text{I}^*$  atoms go over into the unexcited state  $^{127}\text{I}$ . The  $^{129}\text{I}^*$  atoms react with the  $\text{Cl}_2$  to form  $^{129}\text{ICl}$  and  $\text{Cl}$ , while the  $^{127}\text{I}$  atoms recombine with the  $\text{CF}_3$  radicals to form  $\text{CF}_3^{127}\text{I}$ . As a result of the described processes,  $\text{ICl}$  molecules greatly enriched with the isotope  $^{129}\text{I}$  accumulate in the volume.

The possibility of selective action of laser radiation on the iodine isotopes is due to the difference between the isotope spectra and to the narrow lasing line on the  $F = 3 \rightarrow F' = 4$  transition ( $\Delta\omega_L < 1.2 \cdot 10^{-3} \text{ cm}^{-1}$  [6]) of the  $\text{R}^{127}\text{I}$ -molecule PDIL.

Figure 1 shows the calculated hyperfine structure of the  $5^2\text{P}_{1/2} \rightarrow 5^2\text{P}_{3/2}$  transition of the  $^{129}\text{I}$  atom, and the experimentally obtained analogous structure of  $^{127}\text{I}$  [7, 8]. For the most intense components of  $F = 3 \rightarrow F' = 4$  of the  $^{127}\text{I}$  atom and  $F = 4 \rightarrow F' = 5$  of the  $^{129}\text{I}$  atom and  $18 \cdot 10^{-3} \text{ cm}^{-1}$  of the  $^{129}\text{I}$  atom, the shift is  $18 \cdot 10^{-3} \text{ cm}^{-1}$ , and the Doppler width is  $\Delta\omega_L = 8 \cdot 10^{-3} \text{ cm}^{-1}$  (at  $T = 300^\circ\text{K}$  and  $P(\text{RI}) \ll 0.03 \text{ atm}$ ). A measure of the selectivity of the laser action is, naturally, the ratio of the cross sections for stimulated emissions of  $^{127}\text{I}^*$  and  $^{129}\text{I}^*$  at the lasing frequency. This ratio depends on the pressure and on the type of gas [9, 10]. Increasing the line width with increasing pressure decreases the selectivity and, by the same token, imposes an upper bound on the range of possible pressures of the

TABLE 1

Reaction	$k^*, k, \text{cm}^3 \cdot \text{sec}^{-1}$	$k^*/k$
$\text{I}^* + \text{Cl}_2 \xrightarrow{k^*} \text{ICl} + \text{Cl}$	$2,1 \cdot 10^{-13}$	$3,5 \cdot 10^7$ [11]
$\text{I} + \text{Cl}_2 \xrightarrow{k} \text{ICl} + \text{Cl}$	$6 \cdot 10^{-21}$	
$\text{I}^* + \text{CF}_3 \xrightarrow{k^*} \text{CF}_3\text{I}$	$< 2 \cdot 10^{-13}$	
$\text{I} + \text{CF}_3 \xrightarrow{k} \text{CF}_3\text{I}$	$0,90 \cdot 10^{-11}$	$< 0,02$ [12]

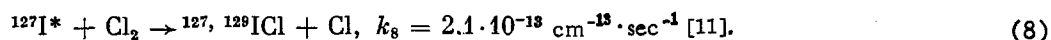
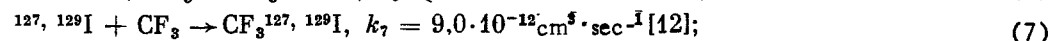
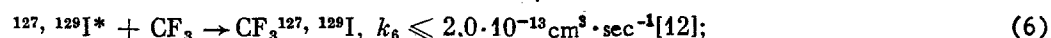
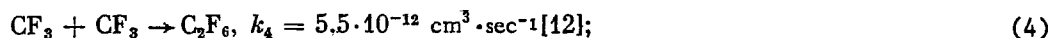
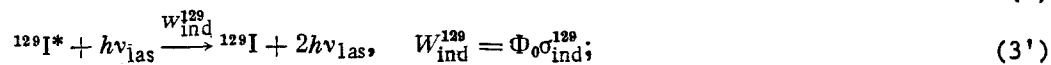
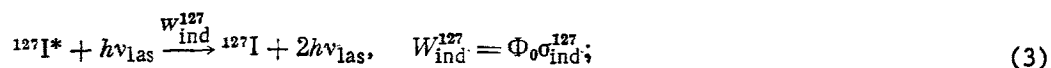
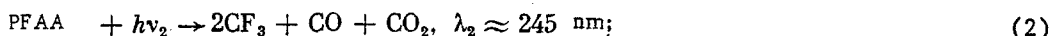
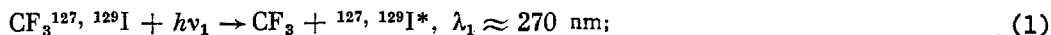
working gases. Thus, at a  $\text{CF}_3\text{I}$  pressure of 0.05 atm we have a ratio  $\sigma_{\text{ind}}^{127}/\sigma_{\text{ind}}^{129} \approx 20$  (at the lasing frequency).

Besides the selectivity of the action of the laser radiation on the iodine isotopes, the isotopes can be separated by using the substantially different chemical activity of the excited  $\text{I}^*(^2\text{P}_{1/2})$  and unexcited  $\text{I}(^2\text{P}_{3/2})$  atoms in the reactions listed in Table 1.

The data in the table show that for the excited  $\text{I}^*(^2\text{P}_{1/2})$  and unexcited  $\text{I}(^2\text{P}_{3/2})$  iodine atoms, the rate constants of the reactions with the molecules  $\text{Cl}_2$  and the radicals  $\text{CF}_3$  differ greatly. Therefore, if the system contains, e.g.,  $\text{Cl}_2$  molecules,  $\text{CF}_3$  radicals, and iodine atoms in the excited and unexcited states, then if the  $\text{Cl}_2$  and  $\text{CF}_3$  are suitably chosen, the excited iodine atoms will go into  $\text{ICl}$  after reacting with  $\text{Cl}_2$ , and the unexcited atoms will go principally into  $\text{CF}_3\text{I}$  after recombining with  $\text{CF}_3$ .

Thus, if a laser field is used to ensure conditions when the  $^{129}\text{I}$  atoms are in the excited state, and the  $^{127}\text{I}$  are not excited, the described reactions will result in  $\text{ICl}$  molecules enriched with the isotope  $^{129}\text{I}$ , and in  $\text{CF}_3\text{I}$  molecules enriched with the isotope  $^{127}\text{I}$ .

We now write down the main processes that occur in the working volume (the source of the  $\text{CF}_3$  radicals was chosen to be perfluoroacetic anhydride (PFAA), i.e., the molecules  $(\text{CF}_3\text{CO})_2\text{O}$ ):



Here  $\Phi_0$  (in photons/ $\text{cm}^2 \cdot \text{sec}$ ) is the flux density of the radiation passing through the volume of the  $\text{R}^{127}\text{I}$  PDIL. The system of differential equations corresponding to the processes (1)-(8) was numerically integrated with a computer to determine the optimal parameters of the enrichment scheme.

The main process that hinders high enrichment is the resonant excitation transfer (5). To suppress the process (5) it is necessary to ensure very rapid binding of the  $^{127}\text{I}(^2\text{P}_{3/2})$  atoms into a  $\text{CF}_3\text{I}$  molecule. This can be done, as shown by calculations, by producing a large excess of  $\text{CF}_3$  radicals compared with the concentration of the  $^{127}\text{I}$  atoms. The concentration of the  $(\text{CF}_3\text{CO})_2\text{O}$  molecules in the initial gas mixture must therefore be maximal. It is bounded from above, however, by the condition that it be possible to resolve the components of the hyperfine structure  $F = 3 \rightarrow F' = 4$  of the  $^{127}\text{I}$  atom and  $F = 4 \rightarrow F' = 5$  of the  $^{129}\text{I}$  atom, and at a ratio  $\sigma_{\text{ind}}^{127}/\sigma_{\text{ind}}^{129} = 20$  it must not exceed  $\sim 10^{18}$  molecules/ $\text{cm}^3$ .

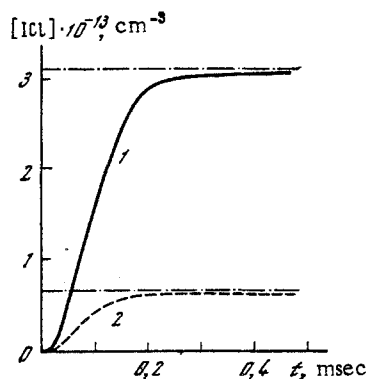


Fig. 2. Calculated time dependences of the concentrations of  $^{129}\text{ICl}$  (1) and  $^{127}\text{ICl}$  (2).

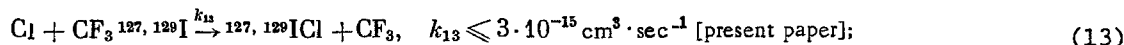
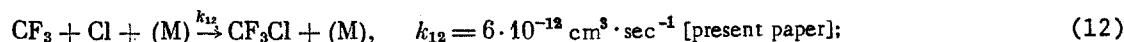
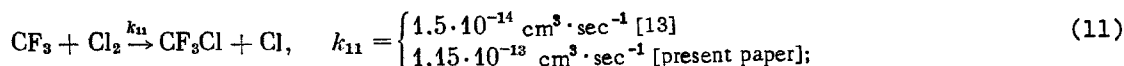
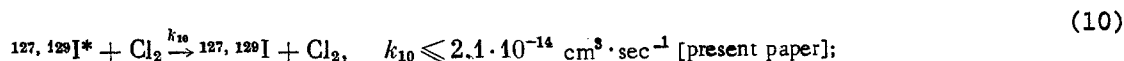
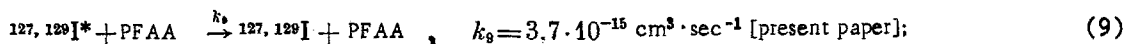
The growth of the gas temperature as a result of photodissociation can cause thermal decomposition of  $\text{CF}_3\text{I}$  and PFAA. The condition  $T < 400^\circ\text{K}$  limits the number of PFAA molecules dissociated per pulse, i.e., the number of  $\text{CF}_3$  radicals produced is  $\sim 10^{16} \text{ cm}^{-3}$ . It follows from the calculations that to obtain an enrichment  $\alpha = 2$  per stage, the number of radicals must be 100 times larger than the number of  $^{127,129}\text{I}$  atoms produced per pulse. Thus, the total number of  $^{127,129}\text{I}$  atoms produced, and consequently the productivity of the enrichment, is  $\sim 10^{14} \text{ atoms/cm}^3$  or  $2.1 \cdot 10^{-8} \text{ g/cm}^3$  of  $^{129}\text{I}$ .

Figure 2 shows the calculated rates of production of the molecules  $^{129}\text{ICl}$  and  $^{127}\text{ICl}$ . The parameters used in the calculations were  $\sigma_{\text{ind}}^{127}/\sigma_{\text{ind}}^{129} = 20$ ,  $[\text{CF}_3^{129}\text{I}]_0/[\text{CF}_3^{127}\text{I}]_0 = 7/3$ , photolyzing pulse duration  $10^{-4} \text{ sec}$ ,  $W_{\text{ind}}^{127} = 10^4 \text{ sec}^{-1}$ , lasing pulse duration  $2 \cdot 10^{-4} \text{ sec}$ ,  $\phi_0 = 3 \cdot 10^3 \text{ W/cm}^2$ ,  $[\text{Cl}_2] = 5 \cdot 10^{16} \text{ cm}^{-3}$ , total number of dissociated  $\text{CF}_3^{127,129}\text{I}$  molecules  $10^{14} \text{ cm}^{-3}$ , number of  $\text{CF}_3$  radicals produced by photodissociation of the PFAA  $10^{16} \text{ cm}^{-3}$ , and enrichment coefficient 2.

The results of the calculations have shown that an increase of the enrichment coefficient per setage to a value  $\alpha = 3-5$  is possible only for a very large (minimum 1000-fold) excess of radicals and this leads, at a fixed total density of the mixture, to a decrease of the yield of the enriched product, which is inversely proportional to the excess of radicals.

## 2. EXPERIMENTAL PART

The experimental investigations of the photolysis of the mixture  $\text{CF}_3^{127}\text{I} + \text{Cl}_2 + \text{PFAA}$  (without a laser field) under conditions close to those given in Sec. 1 have shown that an additional buildup of a considerable amount of  $\text{ICl}$  molecules takes place in this system. The kinetics of the buildup is not described by the reaction scheme (1)-(8). It was found necessary to include into consideration the following additional reactions:



To determine the rate constants of the reactions (9)-(13), we investigated the photolysis of the following mixtures:  $\text{CF}_3^{127}\text{I} + \text{PFAA}$ ;  $\text{PFAA} + \text{Cl}_2$ ; and  $\text{CF}_3\text{I} + \text{Cl}_2$ . The only published value was the rate constant of reaction (11),  $k_{11} = 1.5 \cdot 10^{-14} \text{ cm}^3 \cdot \text{sec}^{-1}$ , but the value of  $k_{11}$  obtained by us turned out to be much larger,  $k_{11} \approx 1.15 \cdot 10^{-13} \text{ cm}^3 \cdot \text{sec}^{-1}$ .

2. To determine  $k_9$ , we used pulsed photolysis of pure  $\text{CF}_3\text{I}$  and of the mixture  $\text{CF}_3\text{I} + \text{PFAA}$ . The initial mixture temperature was  $T_0 \approx 290^\circ\text{K}$ . The pressure of the  $\text{CF}_3\text{I}$  was 0.07 atm, and that of the PFAA was 0.007 atm. The degree of decomposition of the  $\text{CF}_3\text{I}$  by light was  $\approx 1.3\%$ . The duration of the photolyzing pulse at its base was  $t_p \approx 250 \mu\text{sec}$ . During the photolysis, the quartz cell ( $l = 24 \text{ cm}$ ,  $d = 1.6 \text{ cm}$ ) with the investigated gases was located in the working channel of a two-beam pulsed setup for the registration, as a function of time, of the absorption spectra in the ultraviolet and in the visible regions of the spectrum ([3]; see also [14]). We experimentally investigated the kinetics of the accumulation of  $\text{I}_2$  by determining the change of the cell transmission at the wavelength  $\lambda = 4650 \text{ \AA}$  in photolysis of pure  $\text{CF}_3\text{I}$  and of its mixture with PFAA. In addition, we measured by iodometric titration the final concentration of  $\text{I}_2$  ( $[\text{I}_2]^\infty$ ). The experimental results are shown in Figs. 3 and 4.

Reduction of the experimental data by the procedure developed in [3] has enabled us to obtain the rate constant of the reaction (9),  $k_9 = 3.7 \cdot 10^{-15} \text{ cm}^3 \cdot \text{sec}^{-1}$ . This value of the constant pertains to conditions when the mixture temperature does not exceed  $T \leq 390^\circ\text{K}$ .

3. Figure 5 shows the measured final concentrations of the molecules  $\text{CF}_3\text{Cl}$  and  $\text{C}_2\text{F}_6$  as functions of the  $\text{Cl}_2$  content in the initial PFAA +  $\text{Cl}_2$  mixtures in the course of their photolysis without a chlorine filter and with a "jacket" filled with  $\text{Cl}_2$ . To determine the constants  $k_{11}$  and  $k_{12}$ , we numerically solved a differential-equation system corresponding to the reactions (4), (11), and (12), with account taken of the photodissociation of the PFAA and  $\text{Cl}_2$  molecules by the radiation from the IFP-5000 xenon flash lamps. The resultant values were  $k_{11} = (1.15 \pm 0.15) \cdot 10^{-13} \text{ cm}^3 \cdot \text{sec}^{-1}$  and  $k_{12} = (6 \pm 3) \cdot 10^{-12} \text{ cm}^3 \cdot \text{sec}^{-1}$ . These values pertain to a temperature  $T = 310 \pm 20^\circ\text{K}$ . The character of the reaction (12) (binary or ternary) was not verified in the present study.

4. In the mixture  $\text{CF}_3\text{I} + \text{Cl}_2 + \text{SF}_6$ , we investigated the kinetics of accumulation of  $\text{ICl}$  molecules with time by determining the change of the transmission of the constant in the absorption band of the  $\text{ICl}$  molecules ( $\lambda_{\text{max}} \approx 4750 \text{ \AA}$ ). The kinetic measurements were performed with the setup described in [3]. The experimental results are shown in Fig. 6. As seen from Fig. 6, the kinetics of  $\text{ICl}$  production in photolysis of the mixture  $\text{CF}_3\text{I} + \text{Cl}_2 + \text{SF}_6$  proceeds through two stages. At first, during the time of pump action, we have for the  $\text{ICl}$  concentration a very rapid growth that can be naturally attributed to the reaction (8). This is followed by a slow growth of the  $\text{ICl}$  concentration, due to the action of the reaction (13). It is important that the  $\text{ICl}$  concentration  $[\text{ICl}]_{t_p}$  at the instant of termination of the pump pulse be equal, at the experimental accuracy, to the number  $N_{\text{I}^*}$  of the excited iodine atoms produced by the photodissociation of the  $\text{CF}_3\text{I}$  molecules. Thus, under these conditions, all the excited iodine atoms have reacted with the  $\text{Cl}_2$  molecules in accordance with reaction (8), a factor of very great importance for isotope separation. The scatter of the experimental values of  $[\text{ICl}]_{t_p}$  and  $N_{\text{I}^*}$  yields an upper-bound ratio  $k_{10}/k_8 \leq 0.1$ .

The slow change of the  $\text{ICl}$  molecule concentration at  $t > t_p$  enables us to determine the upper limit of the constant  $k_{13}$  at  $T = 300^\circ\text{K}$ :  $k_{13} \leq 3 \cdot 10^{-15} \text{ cm}^3 \cdot \text{sec}^{-1}$ .

5. Thus, our experiments have shown that the scheme of reactions (1-8), which determine the effectiveness of laser separation of iodine isotopes, should be supplemented by reactions (10)-(13). It is easily seen that the reactions (10)-(13) can adversely affect the iodine-isotope enrichment parameters for the following reasons:

1) quenching of  $^{129}\text{I}^*$  in reaction (9) followed by their binding into the molecules  $\text{CF}_3^{129}\text{I}$ ;

2) decrease of the concentration of the  $\text{CF}_3$  radicals in the system on account of the reactions (11) and (12);

3) buildup, in the reaction (13), of  $\text{ICl}$  molecules with an isotope ratio equal to the initial one in the  $\text{CF}_3\text{I}$  molecules.

The experimentally obtained values of the rate constants of reactions (9)-(13) show that the most effective among them, under the conditions necessary for laser isotope separation, are reactions (9) and (13). The most natural method of decreasing the efficiencies of the considered reactions is to decrease the initial concentration of the PFAA and  $\text{CF}_3\text{I}$ . However, a decrease of  $[\text{CF}_3\text{I}]_0$  and  $[\text{PFAA}]_0$  at a fixed pump intensity would lead to a pro-

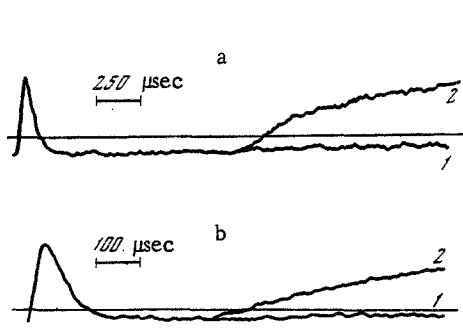


Fig. 3

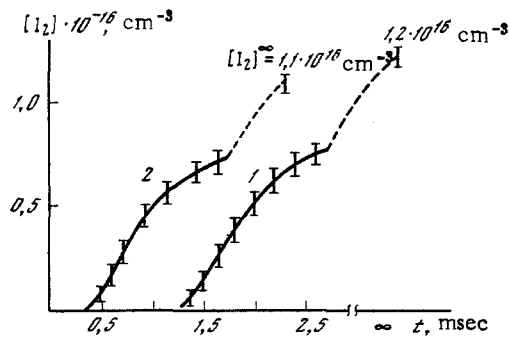


Fig. 4

Fig. 3. Oscillograms showing the change in the transmission of the cell with the investigated gases at the wavelength  $\lambda = 4650 \text{ \AA}$ . 1) Difference between the signals in the comparison channel and in the working channel prior to the photolysis; 2) after the photolysis. a) Photolysis of pure  $\text{CF}_3\text{I}$ ,  $P = 0.07 \text{ atm}$ ; b) photolysis of the mixture  $\text{CF}_3\text{I}$  ( $0.07 \text{ atm}$ ) + PFAA ( $0.007 \text{ atm}$ ). The signal of scattered light from the photolytic lamps can be seen at the beginning of each oscillogram.

Fig. 4. Kinetics of accumulation of molecular iodine in pulsed photolysis of  $\text{CF}_3\text{I}$  (1) and of its mixture with PFAA (2).  $P_{\text{CF}_3\text{I}} = 0.07 \text{ atm}$ ,  $P_{\text{PFAA}} = 0.007 \text{ atm}$ .

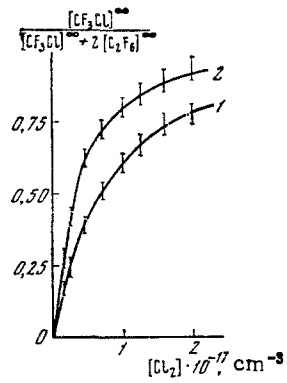


Fig. 5

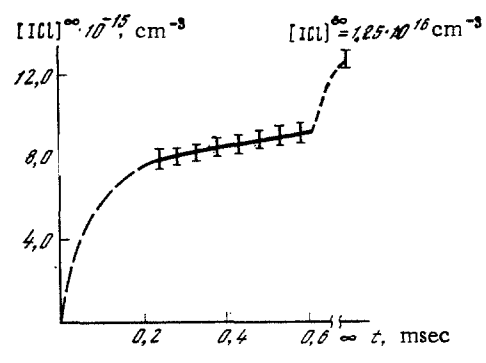


Fig. 6

Fig. 5. Dependence of the ratio  $\frac{[\text{CF}_3\text{Cl}]^\infty}{[\text{CF}_3\text{Cl}]^\infty + 2[\text{C}_2\text{F}_6]^\infty}$  on the initial concentration of the  $\text{Cl}_2$  in pulsed photolysis of a PFAA +  $\text{Cl}_2$  mixture. 1) Photolysis with  $\text{Cl}_2$  filter, 2) photolysis without a filter.

Fig. 6. Kinetics of accumulation of  $\text{ICl}$  molecules in pulsed photolysis of a mixture  $\text{CF}_3\text{I}$  ( $0.0075 \text{ atm}$ ) +  $\text{Cl}_2$  ( $0.04 \text{ atm}$ ) +  $\text{SF}_6$  ( $0.3 \text{ atm}$ ).

portional decrease of the yield of the  $\text{ICl}$  molecules in the laser separation of iodine isotopes. The separation of  $^{127}\text{I}$  and  $^{129}\text{I}$  by this method can remain equally productive despite the decrease of the initial concentrations of the  $\text{CF}_3\text{I}$  and PFAA. For this purpose, it is necessary to increase the pump intensity in such a way as to retain the number of excited iodine atoms and  $\text{CF}_3$  radicals in the system. To prevent the heating that accompanies the increase of the pump energy, the initial  $\text{CF}_3\text{I} + \text{Cl}_2 + \text{PFAA}$  mixture can be diluted with a buffer gas having a large heat capacity and weakly quenching the excited state  $\text{I}^*(^2\text{P}_{1/2})$  of the iodine atom.

Such a gas can be  $\text{C}_5\text{F}_{12}$ , since the quenching constant of the  $\text{I}^*$  by  $\text{C}_5\text{F}_{12}$  molecules, determined in the present study (in analogy with the value of  $k_9$ ) is small,  $k_q \approx 2.5 \cdot 10^{-16} \text{ cm}^3 \cdot \text{sec}^{-1} \ll k_9$ , and the specific heat is quite high,  $(c_{V,300^\circ})_{\text{C}_5\text{F}_{12}} \approx 50 \text{ cal/mole} \cdot \text{deg} \approx 1.3 (c_{V,300^\circ})_{\text{PFAA}}$ .

Thus, it can be assumed if the  $\text{CF}_3\text{I} + \text{PFAA} + \text{Cl}_2 + \text{C}_5\text{F}_{12}$  mixture has parameters  $[\text{CF}_3\text{I}]_0 = 10^{15} \text{ cm}^{-3}$ ,  $[\text{PFAA}]_0 = 2 \cdot 10^{17} \text{ cm}^{-3}$ ;  $[\text{Cl}_2]_0 = 5 \cdot 10^{16} \text{ cm}^{-3}$ ; and  $[\text{C}_5\text{F}_{12}]_0 \sim 10^{18} \text{ cm}^{-3}$ , the harmful processes (9)-(13) will not influence the kinetics of the accumulation of the molecules  $^{127}\text{ICl}$  and  $^{129}\text{ICl}$ , and the enrichment parameters in laser isotope separation will be close to those given in Sec. 1. Preparations are being made at present for a control experiment that should ensure the production of  $\sim 10^{-8} \text{ g/cm}^3$  of the  $^{129}\text{I}$  isotope at an enrichment coefficient  $\alpha \approx 2$ .

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