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Experimental study of the drift motion of SF_6 molecules under the action of CO_2-laser radiation

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Experiments were continued to study the physical nature of the drift motion of SF_6 molecules under the action of radiation from a repetitively pulsed CO_2-laser. To explain the physical nature of the effect, such phenomena as a change in the transport cross-section during laser excitation of molecules, the phenomenon of optical tweezers, thermal diffusion are considered. However, the most reasonable physical explanation of the nature of this effect is the fractionation of a gas mixture as a result of gas dynamic diffusion at the front of a cyclically expanding gas (a variant of barodiffusion).

Keywords: fractionation of gas mixture, thermal diffusion, optical tweezers, barodiffusion.

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Introduction

In papers [1–4] the phenomenon of light-induced drift of atoms and light-weight molecules under the action of cw laser radiation was discovered and studied. The essence of the phenomenon is that when setting the wavelength of the laser on the red (or blue) side of the Doppler broadened absorption line of atoms or molecules their directed diffusion is observed towards (or after) the laser beam. The reason for this is that when a photon is absorbed, the transport cross-section of atoms or molecules increases, and, accordingly, the rate of their directed diffusion decreases.

Similar experiments were carried out in paper [5] to study the drift of sulfur hexafluoride molecules under the action of radiation from repetitively pulsed CO_2 laser. A significant effect was found. Moreover, the SF_6 molecules always moved towards the laser beam, regardless of the wavelength of this radiation. The physical nature of this phenomenon remained unclear.

The nature of absorption of pulsed CO_2-laser radiation by large heavy molecules (such as SF_6) differs significantly from the nature of its absorption by light-weight molecules NH_3 and CH_3F. In large, heavy molecules the main role in the pulsed radiation absorption is played by the broad Lorentz wings of lines of absorption lines [6]. In SF_6 the natural width of these line wings of lines is about $4.5\,cm^{-1}$. In this case, the Doppler shift plays no role.

The transport cross-section increasing during vibrational excitation of SF_6 molecules probably takes place. And this could explain the molecules concentration at the entrance to the cell, where there is the highest radiation intensity. However, this directly contradicts to the authors' statement [5] that the effect was completely out when radiation from cw CO_2-laser of comparable average power was used. This fact indicates that the intensity of laser radiation plays a very important role in this case.

Approximately at the same time, the so-called optical tweezer effect was discovered and widely used [7–9]. Here, the cw laser radiation is focused by a short-focus lens, and atoms, molecules, and particles are captured and retained in its caustic. The effect takes place in vacuum, gas and even liquid. The explanation of the physical nature of the effect based on classical concepts is that atoms and molecules can be polarized. And in the region of a large gradient of laser radiation intensity a force appears that draws atoms and molecules into the region of the highest radiation intensity.

An important feature of the optical tweezer effect is the requirement for high intensity of laser radiation. This property is in complete agreement with the observed dependence of SF_6 molecular drift effect on the use of pulsed or cw CO_2-laser radiation. In the present paper we continue the experiments started in the paper [5] in order to determine the possible role of the optical tweezer effect there. We were interested in the motion of molecules not along the laser beam, but across it.

1. Experimental part

Experimental setup is presented in Fig. 1. The cell was a steel tube with an inner diameter of 14 mm and a length of 500 mm with glued NaCl windows. A tube with an inner diameter of 10 mm and a length of about 70 mm was welded near the inlet window of the cell, the tube served as a ballast volume of about 7% of the total volume of the cell. Through the leaks the gas mixture entered the quadrupole mass-spectrometer, and the gas composition in the ballast volume was recorded in real time by a computer.

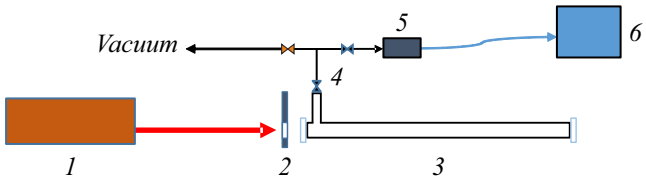

Figure 1. Scheme of the experimental setup: *1* — CO_2-laser, *2* — diaphragm with a diameter of 14 mm, *3* — cell, *4* — leaks, *5* — quadrupole mass-spectrometer, *6* — computer.

A repetitively pulsed CO_2-laser with a nonselective resonator and generation on the 10P20 line was used. When using a gas mixture with the composition CO_2:N_2:He = 2:1:5 and a pressure of 0.5 atm the strongly modulated laser pulse had a duration of about 100 ns at half maximum and a low-intensity tail with duration of about 0.3 μs, which contained about 30% of the energy (Fig. 2, *a*). For comparison, in some experiments, a long laser pulse was used (gas mixture with composition CO_2:N_2:He = 1:4:4 and pressure 0.2 atm (Fig. 2, *b*)). In this case, the pulse peak contained less than 10% of the energy. Accordingly, the low-intensity tail of the pulse with total duration of 10 μs contained more than 90% of the energy.

An unfocused laser beam with a radiation energy density of about 100 mJ/cm^2 was used. The pulse repetition rate was usually 50 Hz. Cw CO_2 laser with a non-selective resonator and a power of about 30 W was also used.

2. Experimental results and discussion

Fig. 3 shows the results of type experiment. When a repetitively pulsed CO_2 laser is switched on, the concentration of SF_6 molecules in the ballast volume decreases, while the concentration of helium atoms increases. After approximately 30 s, a stationary state is reached. Thus, we see that SF_6 molecules are drawn into the laser beam, while helium atoms are pushed out of it. The long pulse (Fig. 2, *b*) use had no effect. At 363 s (Fig. 3) the mass-spectrometer leak was closed, and further readings demonstrate the background of the device.

Fig. 4 shows the experimental results similar to those shown in Fig. 3 using radiation from cw CO_2-laser. Radiation with power of about 20 W was delivered inside the cell. This is almost by three times greater than the average radiation power of the repetitively pulsed CO_2-laser at 50 Hz. Besides, cw laser radiation is absorbed much better than pulsed laser radiation. Thus, at the gas mixture pressure of 8 mm Hg, cw laser radiation (in contrast to pulsed laser radiation) was completely absorbed in the cell. Thus, when the energy input from the cw laser is almost by an order of magnitude greater than from the repetitively pulsed one, the effect of drawing the SF_6 molecules into the laser beam was completely absent. On the contrary, a small ($\sim$ 10%) signal increasing from SF_6 molecules was observed. This can be a consequence of both thermal diffusion and a consequence of gas pressure increasing in the cell due to its heating. The input part of the cell was heated up in 2 min to 60–70°C by cw laser radiation.

The results in Fig. 3 are similar in appearance to the occurrence of the optical tweezer effect. However, other results do not comply with this hypothesis. First of all, this is the dependence of the effect on the gas mixture pressure.

Fig. 5 shows the dependence of the degree of concentration decreasing of SF_6 molecules in the ballast volume on the pressure of gas mixture with the composition SF_6:He = 1:10. Under these conditions the effect reached its maximum value at mixture pressure of 8–10 mm Hg. The physical meaning of this dependence is not clear.

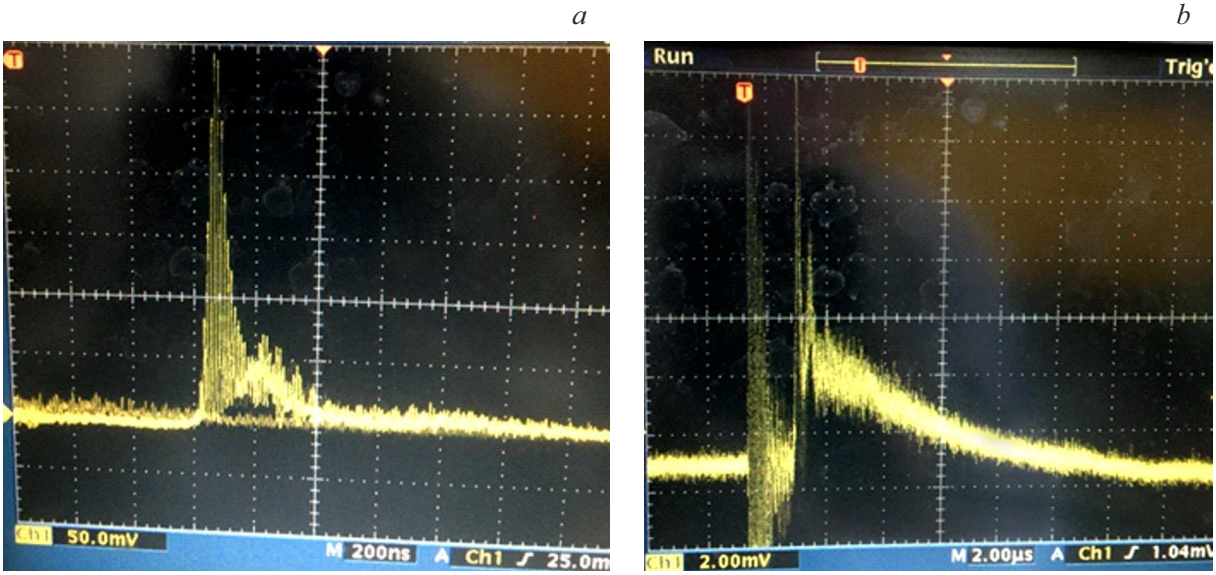

Figure 2. Shape of CO_2-laser pulses.

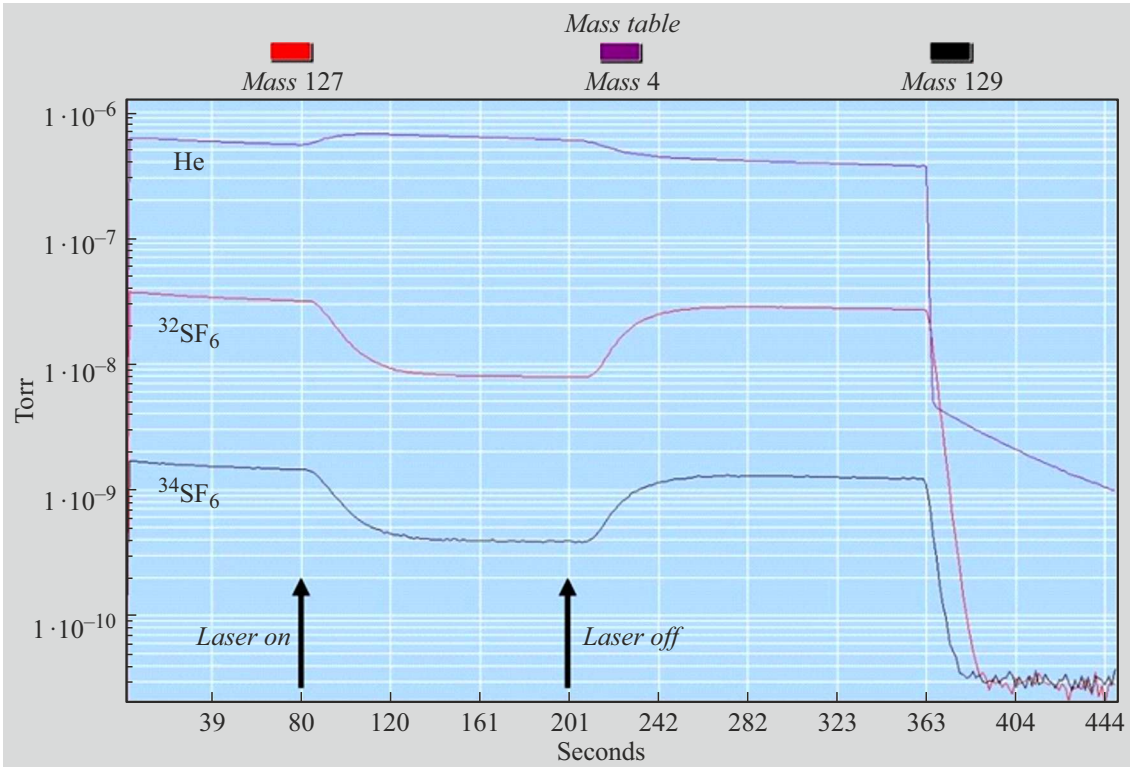

Figure 3. Real-time mass-spectrometer readings during irradiation of a gas mixture SF_6:He = 1:10 and at pressure of 8 mm Hg by radiation of repetitively pulsed CO_2-laser with pulse repetition rate of 50 Hz.

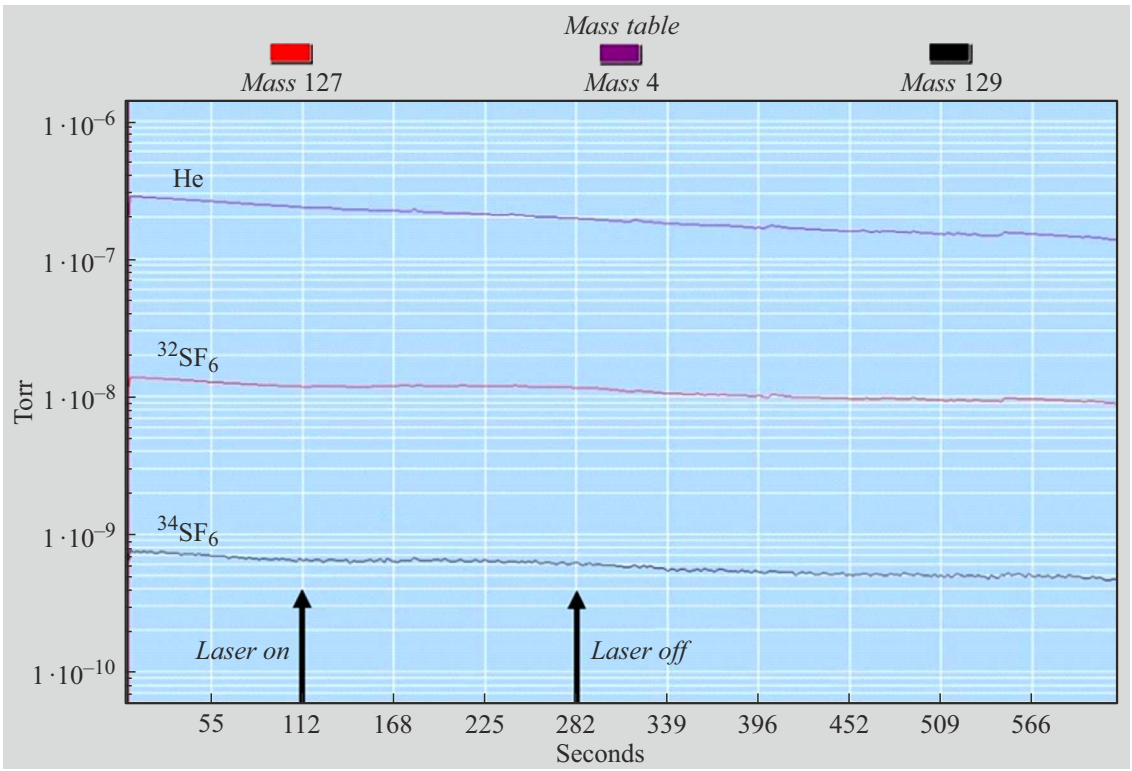

Figure 4. Real-time mass-spectrometer readings during irradiation of gas mixture SF_6:He = 1:10 and at pressure of 8 mm Hg by radiation of cw CO_2-laser.

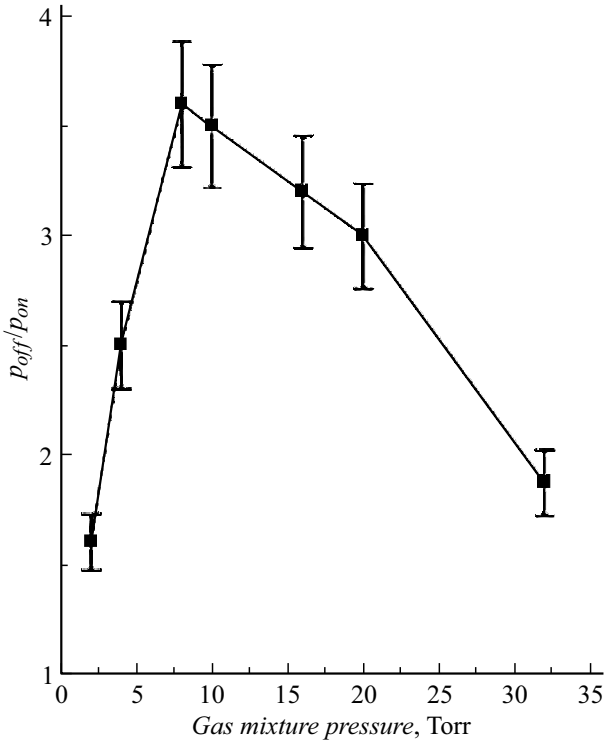

Figure 5. Degree of concentration decreasing of SF_6 molecules in the ballast volume vs. gas mixture pressure when irradiated with radiation from repetitively pulsed CO_2-laser with pulse repetition rate of 50 Hz.

Fig. 6 shows experimental results similar to those shown in Fig. 3 using gas mixture SF_6:He = 1:1 and pressure of 4 mm Hg. Here the effect is much smaller, while it is directed in the opposite direction!

Fig. 7 shows the results of the experiment with gas mixture SF_6:Kr:He = 1:2:8 at total pressure of 11 mm Hg. It can be seen from the Figure that heavy sulfur hexafluoride molecules and heavy krypton atoms are drawn into the laser beam, while light-weight helium atoms are pushed out of it. Similar experiments were performed with a mixture SF_6:Kr = 1:10 at total mixture pressure of 10 mm Hg. The result is similar to the result in Fig. 3, but the degree of the concentration decreasing of SF_6 in the buffer volume was smaller and amounted to about 40%.

The data in Fig. 7 are very similar to the result of thermal diffusion. In this case only the cold area should be in the irradiated volume. Hypothetically, such cooling is possible. It is known that under certain conditions it is possible to cool atoms and molecules with the help of laser radiation.

In order to test this hypothetical possibility of thermal diffusion during cooling of the irradiated volume, we carried out a series of experiments to monitor the stationary temperature of the gas mixtures used. For this a separate steel cell with inner diameter of 36 mm, length of 140 mm with four windows was used. A thin chromel — copel thermocouple (from PMT-2 thermocouple lamp) was introduced into the center of the laser beam. The time for establishing the

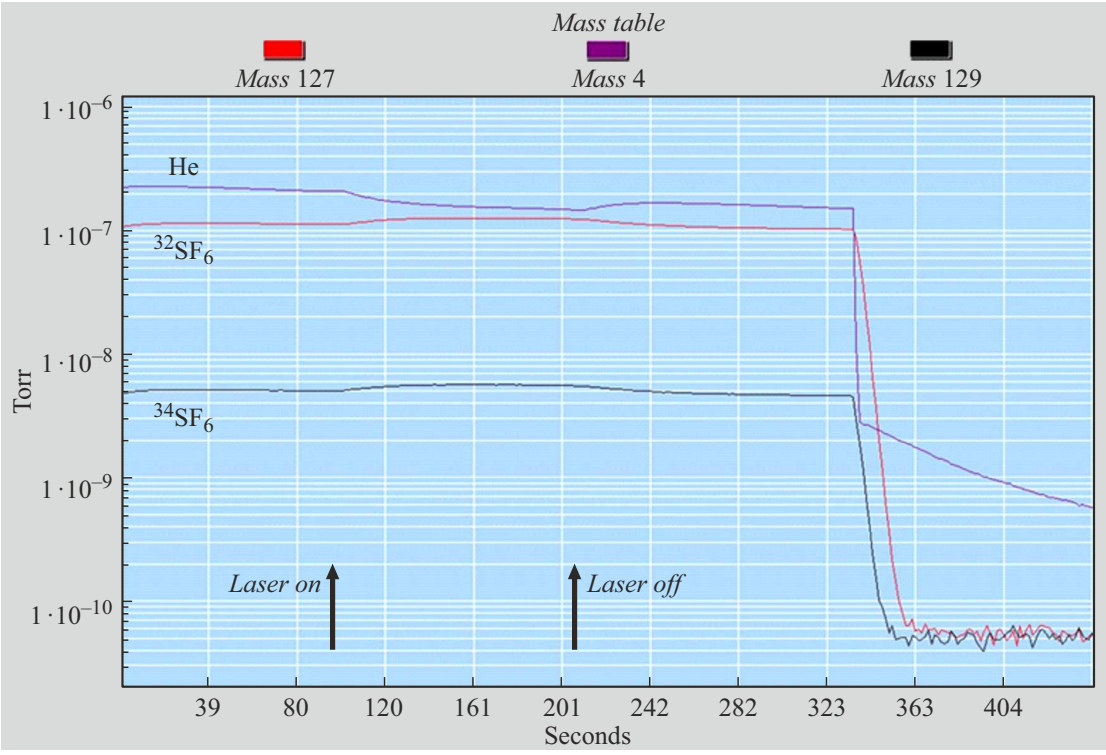

Figure 6. Real-time mass-spectrometer readings during irradiation of a gas mixture SF_6:He = 1:1 and at pressure of 4 mm Hg by radiation of repetitively pulsed CO_2-laser with pulse repetition rate of 50 Hz.

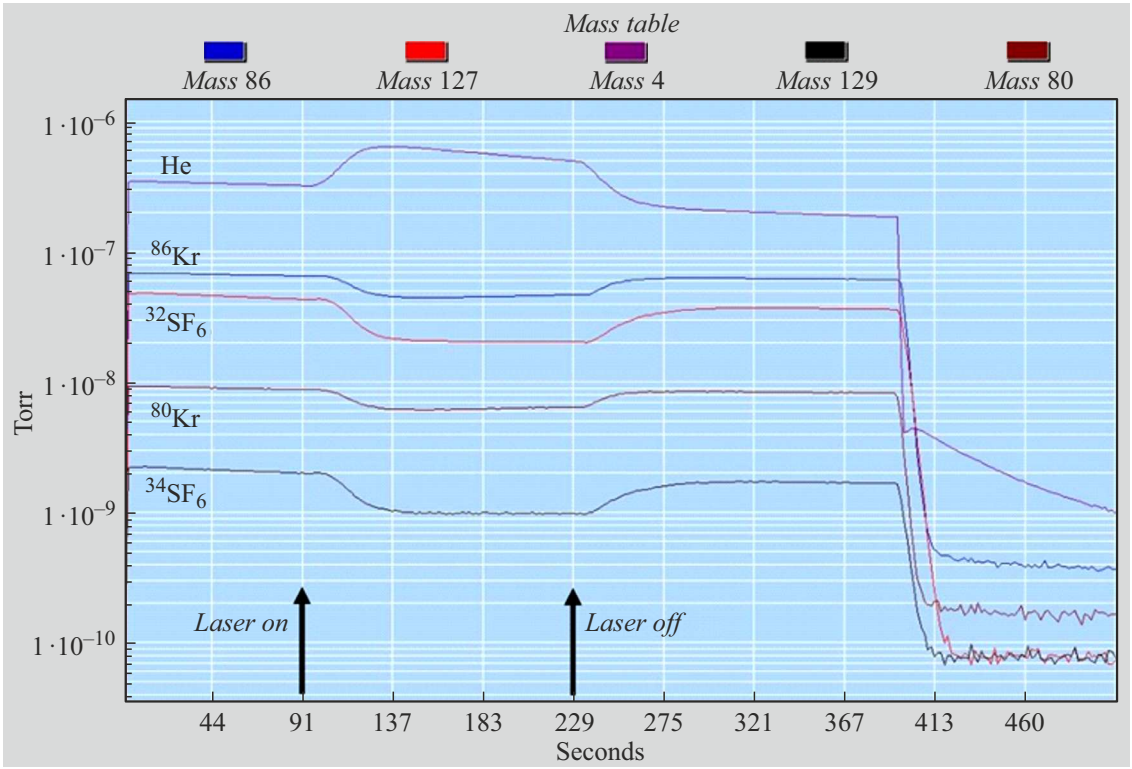

Figure 7. Readings of the mass-spectrometer in real time during the irradiation of the gas mixture SF_6:Kr:He = 1:2:8 at total pressure of 11 mm Hg by radiation of repetitively pulsed CO_2-laser with pulse repetition rate of 50 Hz.

stationary temperature was 1–2 s (the pulse repetition rate was 50 Hz). Fig. 8 shows this stationary temperature vs. the pressure and composition of gas mixtures.

The dependence *1* characterizes the direct heating of the thermocouple by laser radiation. We were interested in any anomalies in the dependences in the pressure region of about 10 mm Hg. However, in the Figure, we see only a monotonic increase in temperature when using mixture SF_6:He = 1:10 (*2*) or monotonic decrease in temperature when helium is added to SF_6 (*3*). This suggests that there is no noticeable hypothetical cooling of the gas mixture.

Note that the effect of drawing resonantly excited atoms into the irradiated region was observed earlier in experiments on light-induced drift of atoms [10]. Here, resonant excited sodium atoms are drawn into the region irradiated by cw laser. The physical explanation of the effect is clear: the transport cross section of the excited atoms increases, and their diffusion slows down. This explanation does not comply with our experiments with sulfur hexafluoride. First, there is no effect when continuous laser radiation is used. Secondly, not only resonant excited molecules ($^{32}SF_6$), but also molecules ($^{34}SF_6$) and atoms (Kr), which do not absorb laser radiation (Fig. 7) are drawn into the radiated area, i.e. not the resonant excitation of molecules is important here, but the size or mass of molecules and atoms.

Thus, neither a possible change in the transport cross-section of excited molecules, nor the phenomenon of optical tweezer, nor thermal diffusion can physically explain the

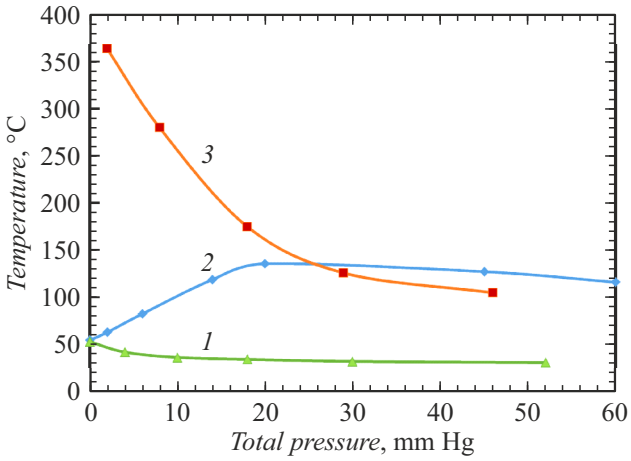

Figure 8. Stationary temperature vs. pressure and composition of gas mixtures: *1* — helium only, *2* — gas mixture SF_6:He = 1:10, *3* — 2 mm Hg SF_6 and helium.

totality of the obtained experimental facts. To understand the nature of the observed effect, the experimental results presented in Fig. 9 are of great importance.

Here we see the transition from the viscous mode of gas mixture flow to the mode of molecular diffusion in a narrow gap of the leak. In the pressure range 450–100 mm Hg the amount of sulfur hexafluoride and helium decreases

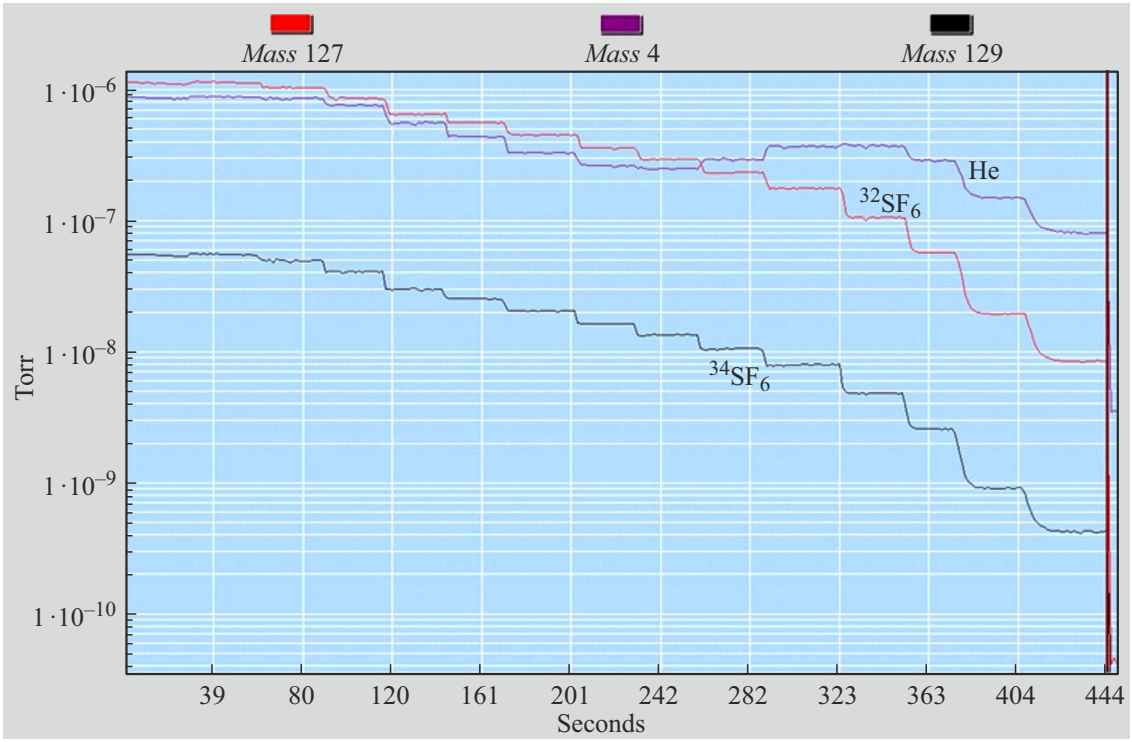

Figure 9. Readings of mass spectrometer in real time with stepwise decrease in the pressure of gas mixture SF_6:He = 1:10 upstream leak from 450 to 2 mm Hg.

downstream the leak in approximately the same way. And in the pressure range 50−2 mm Hg a significant fragmentation of the gas mixture is observed. The change in the relative concentrations of the components is about 10 times. In the mode of molecular diffusion in the leak capillary the helium atoms diffuse from the high-pressure region to the low-pressure region much faster than sulfur hexafluoride molecules. A similar dependence is also observed for the mixture with the composition SF_6:He = 1:1.

A similar situation exists when the gas mixture exits the nozzle into vacuum. Light-weight atoms or molecules leave the central part of the jet faster than heavy ones.

A similar situation is probably realized in the present experiments with the drift of SF_6 molecules. Absorption of pulsed laser radiation leads to rapid heating of the irradiated region of the gas and pressure rise in it. The gas begins to expand into the region of the ballast volume, where there is no radiation, and the pressure is lower. At the boundary of the expanding gas the properties of the so-called barodiffusion appear [11–13]. This is followed by a stage of slow cooling of the irradiated region of the gas and pressure decreasing in it. If the rate of barodiffusion at the boundary of the expanding gas depends nonlinearly on the pressure difference between two regions of the gas, then the existence of such sophisticated dependence of the effect on the pressure and composition of the gas mixture, which is observed in our experiments, is quite possible.

The detailed study and mathematical modeling of barodiffusion under conditions of cyclic laser action on the gas mixture was not the aim of this paper. We only looked for possible evidences of the optical tweezer phenomenon presence here, but did not find them.

We were also interested in the possibility of laser separation of isotopes. With the accuracy of our mass-spectrometer (±0.5%) we did not detect any change in the isotopic composition of SF_6 molecules in the ballast volume. No wonder. The process of separating the components of the gas mixture occurs in the diffusion mode. This is a slow process, and the initial isotopic selectivity of laser excitation of molecules is completely lost here. The isotopic selectivity of molecular diffusion itself is low.

Conclusion

In order to explain the physical nature of the effect, a series of experiments was carried out to study the drift motion of SF_6 molecules in the radiation field of the repetitively pulsed CO_2-laser. The results obtained cannot be explained either by a change in the transport cross-section of molecules upon their laser excitation, or by the optical tweezer phenomenon, or by thermal diffusion. The most appropriate physical explanation of this effect nature is recognized as the fractionation of gas mixture as a result of gas-dynamic diffusion at the front of cyclically expanding gas (barodiffusion option).

Acknowledgments

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Conflict of interest

The authors declare that they have no conflict of interest.

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