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OPTICS
AND LASER PHYSICS

Increase in the Efficiency of the Isotope-Selective Infrared Laser Multiphoton Dissociation of $^{11}BCl_3$ Molecules in a Mixture with SF_6 Serving As a Sensitizer and an Acceptor of Radicals

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A strong increase in the efficiency of the isotope-selective infrared laser multiphoton dissociation of $^{11}BCl_3$ molecules in the natural mixture with $^{10}BCl_3$ by radiation of a pulsed CO_2 laser in the case of admixture of SF_6 molecules, which serve as a sensitizer and simultaneously acceptors of radicals, Cl atoms formed in the dissociation of BCl_3 molecules, has been detected. The yield and selectivity of dissociation of $^{11}BCl_3$ molecules increase by several times and the threshold energy density for the dissociation of molecules decreases significantly in the case of their irradiation in the mixture with SF_6 compared to irradiation without SF_6. This property allows the single-frequency isotope-selective dissociation of $^{11}BCl_3$ molecules by unfocused laser radiation at a moderate energy density (≈3–5 J/cm^2), which is important and relevant for the practical implementation of the laser separation of boron isotopes.

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1. INTRODUCTION

Interest in the laser separation of boron isotopes has increased recently [1–4] because of their application in important industrial branches, in medicine [5–7], and in space experiments [8]. Boron in nature exists in the form of two isotopes ^{10}B (about 19.8%) and ^{11}B (about 80.2%) [5, 6]. The thermal neutron capture cross section of ^{10}B is very large, about 3837 b [9] (1 b = 10^{-24} cm^2). This cross section for most nuclides is about several or fractions of barn. For this reason, materials enriched in the ^{10}B isotope (boric acid, boron carbide, and other compounds) are widely used in the nuclear power industry. The ^{11}B isotope is used in electronics industry as a dopant in the manufacture of semiconductor products [6, 7]. Consequently, the development of methods of efficient technologies for the separation of boron isotopes is important and relevant.

The most efficient method of molecular laser separation of isotopes is currently the selective infrared multiphoton dissociation of molecules by the radiation of a pulsed periodic CO_2 laser [10, 11]. Therefore, it is reasonable to use this method in the technological process of separation of boron isotopes. It was successfully applied in the practical technology for laser separation of carbon isotopes [12, 13].

For the molecular laser separation of isotopes with existing efficient pulsed CO_2 lasers, infrared absorption bands of the chosen molecules should be in the generation range of the CO_2 laser and have a fairly large (≥5–10 cm^{-1}) isotopic shift. In addition, a quite low energy density of laser radiation for the efficient isotope-selective dissociation of these molecules is desired.

One of the most suitable compounds for the laser separation of boron isotopes is the gaseous BCl_3 compound. Infrared absorption bands of ν_3 vibrations of $^{11}BCl_3$ (≈954.2 cm^{-1}) and $^{10}BCl_3$ (≈993.7 cm^{-1}) molecules [14] are in resonance with the *P* and *R* branches of the 10.6-μm band of the CO_2 laser, respectively. The isotopic shift between the indicated infrared absorption bands of $^{11}BCl_3$ and $^{10}BCl_3$ molecules is $\Delta\nu_{is} \approx 39.5$ cm^{-1} [14]. The isotope-selective laser infrared multiphoton dissociation of molecules was demonstrated for the first time just with BCl_3 molecules [15]. The isotope-selective dissociation of BCl_3 molecules in the radiation of a high-power pulsed CO_2 laser with the use of various acceptors of radicals (H_2, NO, H_2S, D_2S, HBr, etc.) to suppress the association of BCl_2 radicals and Cl atoms formed at the dissociation of BCl_3 molecules [16] was studied in detail in numerous subsequent works [16–26].

It was established that oxygen is a quite good acceptor of radicals to achieve a high selectivity and a high dissociation yield for BCl_3 molecules [17–19, 22]. The final products of the infrared multiphoton dissociation of BCl_3 molecules in the presence of oxygen are B_2O_3 and Cl_2 [17]. The solid B_2O_3 compound is deposited in the form of a film on the walls and windows of the cell, where the gas is irradiated. It was shown in [17, 23] that acceptors of radicals are hardly responsible for any significant increase in the selectivity and yield of dissociation of BCl_3, as well as for the reduction of the threshold for the dissociation of molecules.

Previous studies show that the efficient infrared multiphoton dissociation of BCl_3 molecules requires pump laser radiation with fairly high energy densities ($\Phi \geq 20$ J/cm^2) [23, 24] mainly because of a high binding energy of the molecules (≈110 kcal/mol [27]). Consequently, only focused laser beams can induce the infrared dissociation of molecules. At the same time, the use of focused laser beams usually reduces the isotopic selectivity of the dissociation of molecules [10, 11].

The selectivity of dissociation of $^{10}BCl_3$ and $^{11}BCl_3$ molecules excited by single-frequency laser radiation is $\alpha(^{10}B/^{11}B) \leq 8$ and $\alpha(^{11}B/^{10}B) \leq 2.5$, respectively [22–24]. In this case, the dissociation yields for molecules (recalculated to the laser-irradiated volume of the gas in the cell) were low ($\beta_{10} \approx \beta_{11} \leq (2–8)\times10^{-4}$ [24]). The selectivity and yields of the dissociation of BCl_3 molecules pumped by two-frequency laser radiation are much higher ($\alpha(^{10}B/^{11}B) \geq 8$; $\beta_{10} \geq 10\%$; $\beta_{11} \geq 20\%$) [28, 29]. In this case, the threshold for the dissociation of molecules decreased significantly (to $\Phi_1, \Phi_2 \geq 2–3$ J/cm^2). However, the practical technological implementation of the laser separation of boron isotopes with the two-frequency dissociation of molecules is very difficult.

In this work, we find that the use of SF_6 molecules as sensitizers and acceptors of radicals in the isotope-selective laser infrared molecule dissociation of BCl_3 molecules significantly increases the efficiency of the dissociation of $^{11}BCl_3$ molecules. This is very important and relevant for the technology of the laser separation of boron isotopes.

2. EXPERIMENT

The experimental setup (Fig. 1) included a frequency-tunable pulsed CO_2 laser, forming optical elements, a 112-mm-long stainless steel gas cell 24.2 cm^3 in volume with BaF_2 windows for the irradiation of studied molecules, and calorimetric (TPI-2-5) and pyroelectric (SensorPhysics Model 510) detectors to measure the energies incident and transmitted

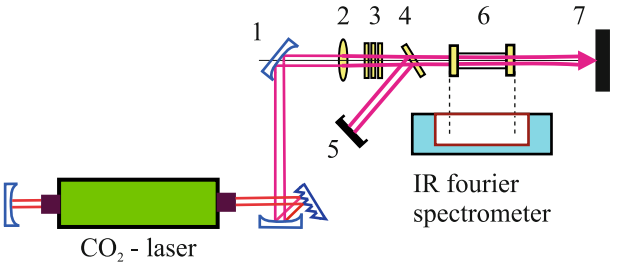

Fig. 1. (Color online) Schematic of the experimental setup: (1) mirror, (2) long-focus lens, (3) attenuators of laser radiation, (4) splitter plate, (5) detector of radiation, (6) cell with the irradiated gas, and (7) absorber of radiation.

through the cell. The pulse of the CO_2 laser consisted of the leading peak with a FWHM duration of about 80 ns and the tail part with a FWHM duration of about 750 ns, which contained approximately one-third of the energy of the pulse. The laser beam was collimated by a long-focus lens ($f = 1$ m) to the cell. The energy in the laser pulse was 0.7 to 3.0 J, depending on the lasing frequency and on the partial composition of the laser mixture. The laser frequency was varied in the range of 9.2–10.8 μm. To determine the frequencies of radiation lines of the CO_2 laser, we used an optoacoustic detector with the NH_3 reference gas. The transverse energy density distribution and the area of the cross section of the laser beam were determined by scanning of a microprobe across the laser beam in the mutually perpendicular directions at the input and output of the cell.

3. METHOD

The infrared multiphoton dissociation of BCl_3 molecules selective in boron isotopes was carried out with the tuning of the frequency of the CO_2 laser to the 932.96-cm^{-1} 10P32 line, which coincides with the long-wavelength wing of the absorption band of the ν_3 vibration of $^{11}BCl_3$ molecules (≈954.2 cm^{-1} [14]). The absorption band of the ν_3 vibration of molecules SF_6 (≈948 cm^{-1} [30]) is redshifted by about 6.2 cm^{-1} from the center of the absorption band of $^{11}BCl_3$ molecules (Figs. 2a–2c). The absorption bands of both $^{11}BCl_3$ and SF_6 molecules under infrared laser multiphoton excitation are redshifted (to the laser frequency) because of the anharmonicity of vibrations and fall in the exact resonance with the laser field [31–34]. The effective infrared multiphoton excitation of both SF_6 and $^{11}BCl_3$ molecules occurs and the absorbed energy is transferred from the former to the latter molecules. Since the absorption bands of SF_6 and $^{11}BCl_3$ molecules almost coincide, the vibrational–vibrational energy exchange between them is resonant and occurs very effectively [35, 36]. This exchange increases the

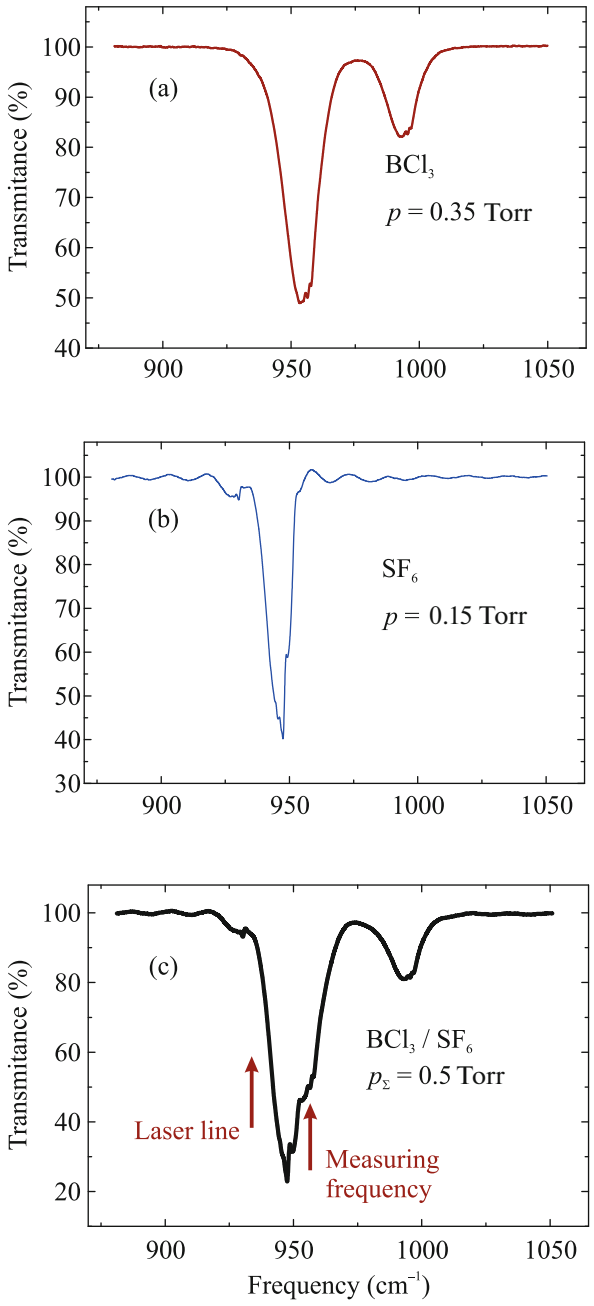

Fig. 2. (Color online) (a) Infrared absorption bands of the ν_3 vibrations of $^{11}BCl_3$ and $^{10}BCl_3$ molecules in the 11.2-cm-long cell at a pressure of 0.35 Torr. (b) Infrared absorption band of the ν_3 vibrations of SF_6 molecules in the cell at a pressure of 0.15 Torr. (c) Infrared absorption bands of the ν_3 vibrations of $^{11}BCl_3$, $^{10}BCl_3$, and SF_6 molecules in the cell at pressures of BCl_3 and SF_6 molecules of 0.35 and 0.15 Torr, respectively.

yield of the dissociation of $^{11}BCl_3$ molecules. Since the dissociation energy of SF_6 molecules (≈92 kcal/mol [37]) is lower than that of BCl_3 molecules, SF_6 molecules are also dissociated at high pump energy densities.

Laser infrared multiphoton dissociation of $^{11}BCl_3$ molecules occurs through the photochemical reaction

$$^{11}BCl_3 + nh\nu \rightarrow {}^{11}BCl_2 + Cl, \quad (1)$$

where $nh\nu$ means n absorbed infrared laser photons.

The products of dissociation induced by the irradiation of the gas were enriched in the ^{11}B isotope, whereas the residual BCl_3 gas was enriched in the ^{10}B isotope. Dissociation yields, which are fractions of molecules dissociated in the irradiated volume per laser pulse, were determined from changes in infrared absorption spectra for each of the BCl_3 isotopic components.

Absorption spectra were recorded with an FT-801 infrared Fourier transform spectrometer. The dissociation yields β_{11} and β_{10} were calculated from the partial pressures of the respective components $^iBCl_3$ (i = 10, 11) before ($p_{i,0}$) and after (p_i) irradiation by laser pulses using the relation

$$\beta_i = \Gamma^{-1}[1 - (p_i/p_{i,0})^{1/N}], \quad (2)$$

where $\Gamma \approx 0.083$ is the ratio of the irradiated volume ($V_{irr} \approx 2.0$ cm^3) to the volume of the cell (V_{cell} = 24.2 cm^3) and N is the number of pump pulses. The intensities of the absorption band of $^{11}BCl_3$ molecules before and after irradiation were measured at a frequency of about 954.2 cm^{-1} (approximately at the maximum of the absorption band of $^{11}BCl_3$ molecules), at which SF_6 molecules hardly contribute to absorption at pressures used in experiments (see Fig. 2b). The selectivity $\alpha(^{11}B/^{10}B)$ was determined as the ratio of the dissociation yields of $^{11}BCl_3$ and $^{10}BCl_3$ molecules

$$\alpha(^{11}B/^{10}B) = \beta_{11}/\beta_{10}. \quad (3)$$

4. RESULTS AND DISCUSSION

The main parameters of selective laser infrared multiphoton dissociation of BCl_3 molecules, namely, the dissociation yields β_{11} and β_{10} of $^{11}BCl_3$ and $^{10}BCl_3$ molecules, respectively, as well as the selectivity $\alpha(^{11}B/^{10}B)$ of the dissociation of $^{11}BCl_3$ molecules from $^{10}BCl_3$ molecules, were measured in the experiments. The dependences of these parameters on the pressures of the irradiated BCl_3 and SF_6 gases, as well as on the pump laser energy density, were obtained.

Figure 3 presents infrared absorption bands of the ν_3 vibrations of $^{11}BCl_3$, $^{10}BCl_3$, and SF_6 molecules (*1*) before and (*2*) after laser irradiation and the infrared absorption bands of the 851.4 cm^{-1} ν_1 vibration and the 909.1 cm^{-1} ν_8 vibration of the formed SF_5Cl product [38]. The initial pressures of BCl_3 and SF_6 molecules in the cell were 0.35 and 0.15 Torr, respectively.

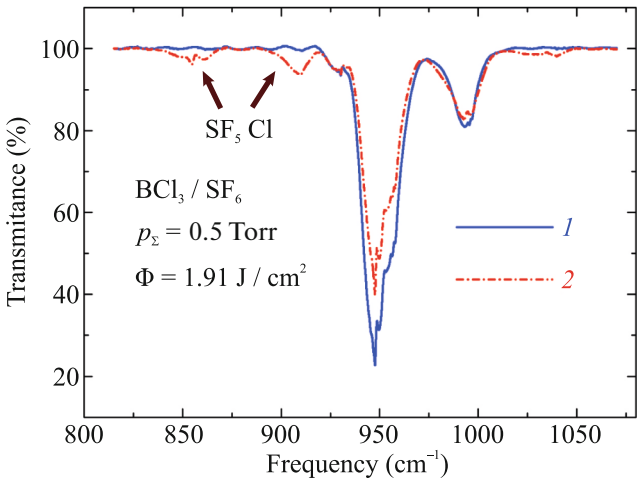

Fig. 3. (Color online) Infrared absorption bands of the ν_3 vibrations of $^{11}BCl_3$, $^{10}BCl_3$, and SF_6 molecules (*1*) before and (*2*) after laser irradiation and the infrared absorption bands of the formed SF_5Cl product with centers at frequencies of 851.4 cm^{-1} (ν_1 vibration) and 909.1 cm^{-1} (ν_8 vibration) [38]. The initial pressures of BCl_3 and SF_6 molecules in the cell are 0.35 and 0.15 Torr, respectively. The energy density of exciting laser radiation is 1.91 J/cm^2 and the number of pulses is $N = 1800$.

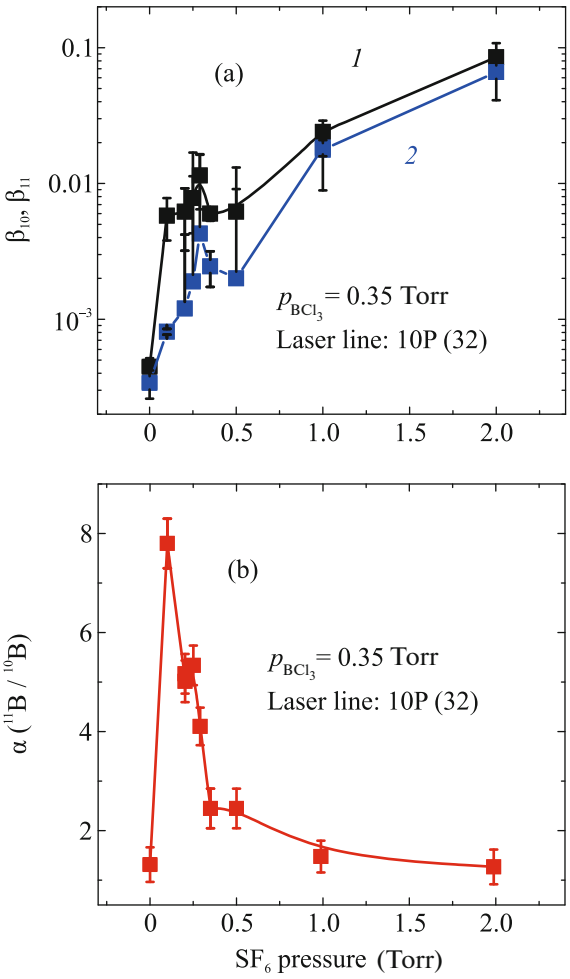

Fig. 4. (Color online) (a) Dissociation yields (*1*) β_{11} and (*2*) β_{10} and (b) the dissociation selectivity $\alpha(^{11}B/^{10}B)$ versus the pressure of CF_6 molecules at the pressure of BCl_3 molecules of 0.35 Torr and a pump laser energy density of 2.1 J/cm^2.

The pump laser energy density was 1.91 J/cm^2. The number of irradiation pulses was $N = 1800$. In addition to SF_5Cl molecules, we detected BCl_2F, $BClF_2$, and BF_3 molecules in the products of dissociation; their infrared absorption spectra are not presented in Fig. 3.

Figure 4 shows the dependences of (a) dissociation yields β_{11} and β_{10} of $^{11}BCl_3$ β_{11} and $^{10}BCl_3$ molecules, respectively, and (b) the selectivity $\alpha(^{11}B/^{10}B)$ of the dissociation of $^{11}BCl_3$ molecules from $^{10}BCl_3$ molecules on the pressure of SF_6 in the range of 0.05–2.0 Torr at a fixed pressure of 0.35 Torr of BCl_3 molecules and the energy density of pump radiation of $\Phi \approx 2.1$ J/cm^2. It is seen that dissociation yields of $^{11}BCl_3$ and $^{10}BCl_3$ molecules in the considered pressure range of SF_6 molecules increase from $\beta_{11} \approx 4.5 \times 10^{-4}$ and $\beta_{10} \approx 3.4 \times 10^{-4}$ at the irradiation of BCl_3 molecules in the absence of SF_6 to $\beta_{11} \approx 8.5 \times 10^{-2}$ and $\beta_{10} \approx 7.2 \times 10^{-2}$, respectively, in the case of the irradiation of BCl_3 molecules in the presence of SF_6 at a pressure of 2.0 Torr.

Thus, the dissociation yield of BCl_3 molecules irradiated in the presence of SF_6 increases by more than two orders of magnitude. It is noteworthy that the addition of SF_6 molecules to a pressure of only 0.1 Torr increases the dissociation yield of $^{11}BCl_3$ molecules by more than an order of magnitude. This also means that the addition of SF_6 molecules strongly reduces the threshold energy density for the dissociation of $^{11}BCl_3$ molecules. The dissociation yields of $^{11}BCl_3$ and $^{10}BCl_3$ molecules increase monotonically with the pressure of SF_6 in the range of ≈0.5–2.0 Torr.

The selectivity $\alpha(^{11}B/^{10}B) \approx 1.3$ of the dissociation of $^{11}BCl_3$ molecules from $^{10}BCl_3$ molecules irradiated in the absence of SF_6 molecules increases sharply to $\alpha(^{11}B/^{10}B) \approx 8.0$ in the presence of SF_6 at a pressure of ≈0.15–0.2 Torr. With a further increase in the pressure of SF_6 to ≈0.4 Torr, the selectivity decreases rapidly to $\alpha(^{11}B/^{10}B) \approx 3.0$ and then decreases slowly to $\alpha(^{11}B/^{10}B) \approx 1.0$ at a pressure of $SF_6 \geq 1.5$ Torr. Thus, the selectivity of the dissociation $\alpha(^{11}B/^{10}B)$ in the case of irradiation of BCl_3 molecules at a pressure of 0.35 Torr in the presence of SF_6 molecules is maximal at the pressure of SF_6 in the range of 0.15–0.25 Torr.

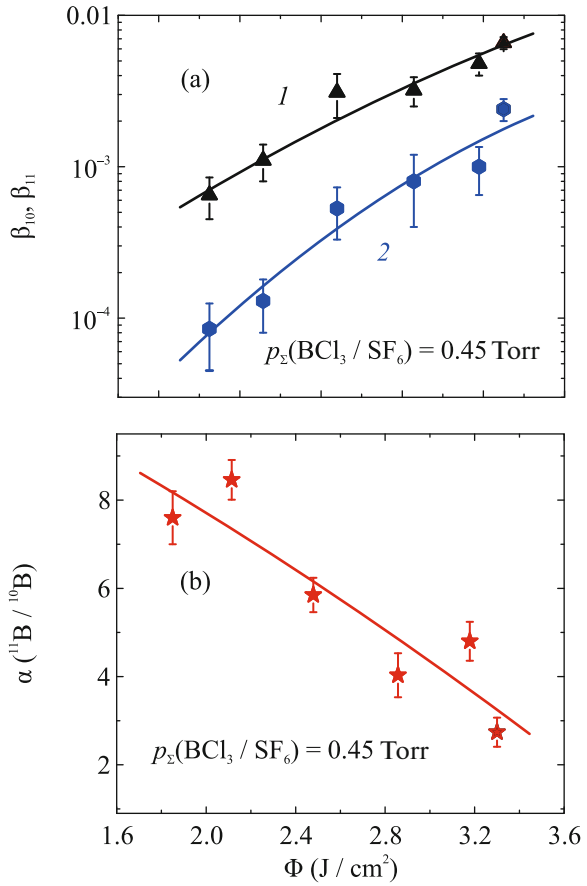

Fig. 5. (Color online) (a) Dissociation yields (*1*) β_{11} and (*2*) β_{10} and (b) the dissociation selectivity $\alpha(^{11}B/^{10}B)$ versus the energy density of exciting laser radiation at the pressures of BCl_3 and SF_6 molecules in the cell of 0.35 and 0.1 Torr, respectively.

The selectivity $\alpha(^{11}B/^{10}B)$ in the presence of SF_6 is more than a factor of 3–5 higher than that for BCl_3 molecules irradiated in the absence of SF_6.

Figure 5 presents the dependences of the dissociation yields of $^{11}BCl_3$ and $^{10}BCl_3$ molecules, as well as the dissociation selectivity $\alpha(^{11}B/^{10}B)$, in the mixture of BCl_3 and SF_6 molecules in the cell at their partial pressures of 0.35 and 0.1 Torr, respectively, on the energy density of exciting laser radiation in the range from ≈1.8 to 3.4 J/cm^2. It is seen that the dissociation yields of BCl_3 molecules increase rapidly with the pump energy density because infrared dissociation at the considered laser energy densities is above-threshold and is comparatively far from saturation in view of a high binding energy of BCl_3 molecules.

On the contrary, the selectivity of the dissociation of $^{11}BCl_3$ molecules $\alpha(^{11}B/^{10}B)$ decreases rapidly with an increase in the pump energy density. At the same time, the dissociation selectivity in the presence of SF_6 molecules at all studied pump energy densities is a factor of 2–5 higher than that in the absence of SF_6. The observed dependences of the dissociation yields and selectivity on the pump energy density are characteristic of the isotope-selective laser infrared multiphoton dissociation for numerous types of molecules [10, 11].

5. CONCLUSIONS

A strong increase in the efficiency of isotope-selective infrared laser multiphoton dissociation of $^{11}BCl_3$ molecules in the natural mixture with $^{10}BCl_3$ has been detected when the irradiated mixture is supplemented with SF_6 molecules, which resonantly absorb laser radiation and serve as sensitizers and acceptors of radicals. It has been shown that the yield and selectivity of dissociation of $^{11}BCl_3$ molecules in the mixture in the presence of SF_6 molecules are one or two orders of magnitude and a factor of 2–5 higher and the threshold energy density for dissociation is about an order of magnitude lower than the respective parameters in the case of the absence of SF_6 molecules. This property allows the single-frequency isotope-selective infrared laser multiphoton dissociation of $^{11}BCl_3$ molecules by unfocused laser beams at a moderate energy density (≤4–5 J/cm^2). The results obtained in this work are important and relevant for the development of a laser technology for separation of boron isotopes because they make it possible to obtain the BCl_3 molecular gas highly enriched in the ^{10}B isotope because of the dissociation of $^{11}BCl_3$ molecules in the natural mixture with $^{10}BCl_3$ molecules.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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