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Features of absorption of intense IR emission by SF_6 molecules

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It is demonstrated experimentally that the frequency of the minimum absorption by SF_6 molecules is shifted into the long-wave region at high intensities of the incident radiation and at high IR pulse durations exceeding the times of rotational and vibrational relaxation. The observed singularities of the absorption spectrum are illustrated with the chemical reaction SF_6+H_2 as an example, and an explanation of the results is presented.

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A study[1] of the selective dissociation of the SF_6 molecule in a strong IR laser field has revealed a frequency shift that brings about a most effective rate of collisionless dissociation of this molecule. We have shown earlier[2] that when the SF_6 molecule is exposed to a strong pulse of CO_2 laser radiation with a pulse duration τ_p exceeding the rotational and vibrational relaxation times ($\tau_{r\text{-}r}$ and $\tau_{v\text{-}v}$), a strong change is observed in the absorption spectrum of the SF_6 molecules when the intensity of the incident radiation is changed. The present paper is devoted to a more detailed study of this phenomenon at various IR intensities.

1. EXPERIMENTAL SETUP

An oscillogram of the irradiating pulse is shown in Fig. 1. The radiation source was a pulsed electric-discharge atmospheric-pressure CO_2 laser similar to that described in[3]. The electrode length was 90 cm, and the distance between electrodes was 5 cm. The laser-radiation frequency selection was effected with a diffraction grating operating in the self-collimation regime. The exit mirror of the laser was an NaCl substrate on which a dielectric coating having a reflection coefficient 40% was sputtered. The working medium was a mixture of the gases CO_2, N_2 and He in a ratio 1 : 1 : 2. The lasing pulse duration (Fig. 1) was ~3 μsec at half-height. The CO_2 laser energy on individual vibrational-rotational transitions reached 6 J.

The cell (Fig. 2) for the investigation of the IR absorption and for the initiation of the chemical reaction in the SF_6+H_2 mixture was 40 cm long. The cell windows were made of NaCl and were inclined at the Brewster angle. The CO_2-laser radiation incident on the cell was collimated into a beam of ~6 mm diameter and practically homogeneous over its cross section (over the entire length of the cell). The waveform of the lasing pulses was registered with a Ge : Au photoresistor ($T_{\text{work}}=77$ K), the signal from which was fed to the input of an S8-2 oscilloscope with a bandwidth up to 8 MHz. The time constant of the radiation receiver was not worse than 200 nsec. The calorimetric energy meters (of the KIM type) ensured a measurement reproducibility

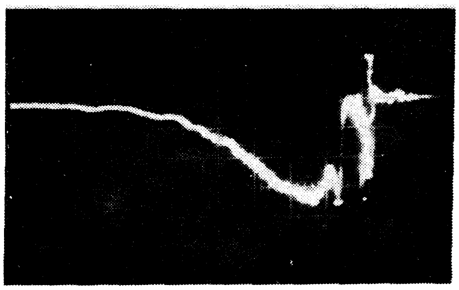

FIG. 1. Oscillogram of CO_2-laser pulse. Time sweep 1 μsec/div.

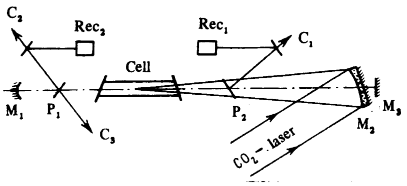

FIG. 2. Diagram of experimental setup: Rec$_1$, Rec$_2$—IR receivers (Ge: Au); C$_1$, C$_2$, C$_3$—calorimeter for the measurement of the incident (C$_1$) and transmitted energy of the CO$_2$ laser (C$_3$) and of the HF generation energy (C$_2$); M$_1$, M$_2$, M$_3$ — mirrors; P$_1$, P$_2$—plane parallel plates of BaF$_2$.

not worse than 2%. The experimental setup is illustrated in Fig. 2.

In the experiments we used sulfur hexafluoride of natural isotopic composition. The table lists the impurity content of the SF$_6$, the total amount of the impurities being not more than ~ 0.6%.

Impurity:	S_2N_4	CF_4	SO_2	SiF_4	CS_2	SOF_2	SO_2F_2	SOF_4
C, %:	0.014	0.466	0.04	0.014	<0.01	<0.01	<0.01	<0.01

2. ABSORPTION OF IR RADIATION BY SF$_6$ MOLECULES

As already mentioned, when SF$_6$ molecules are exposed to intense pulsed CO$_2$-laser radiation, a change is observed in the absorption spectrum. We have shown earlier[2] that at high radiation intensities (up to 10^7 W/cm^2) and at a radiation-pulse duration longer than the time of rotational relaxation and v-v exchange, the maximum absorption of the ν_3 band of the SF$_6$ molecule is shifted by ~ 14 cm^{-1} towards the red end away from the frequency $\nu = 948$ cm^{-1} corresponding to the maximum absorption of low-intensity IR radiation by the ν_3 mode of the SF$_6$ molecule.[4]

Figure 3 shows a plot of E_{abs}/E_{inc} against the CO$_2$ emission frequency. The SF$_6$ pressure was 2.7 Torr, and the incident-radiation power was ~ 10^7 W/cm^2. For comparison, the same figure shows the spectrum of the linear absorption of ^{32}SF$_6$ recorded with a high-resolution instrument.[5] It follows from these data that the maximum absorption is observed at the frequencies 932–935 cm^{-1} the P_{30}–P_{32} emission lines of the CO$_2$ laser), which are close to the ν_3 absorption band of the ^{34}SF$_6$ molecule (the isotopic shift for ^{34}SF$_6$ is 17.4 cm^{-1} [4]). In this spectral interval, the value of the absorbed energy is 1.6 times larger than the energy absorbed at the frequency corresponding to the maximum of the linear absorption of ^{32}SF$_6$. This is more than 30 times larger than the ratio of the energy absorbed at the frequencies ~ 934 and ~ 948 cm^{-1} at low irradiation intensities (when Bouguer's law holds).

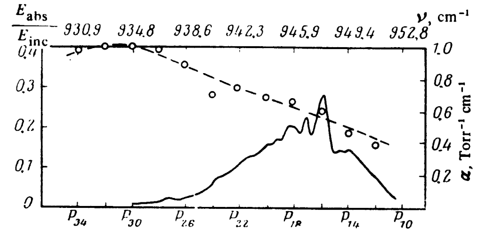

FIG. 3. Ratio of the absorbed energy to the incident energy as a function of the frequency of the incident radiation—dashed curve (concentration SF$_6$: H$_2$ = 2.5 : 1, total pressure 4 Torr; cell length 40 cm). The absorption band of the ν_3 mode of ^{32}SF$_6$[5] is shown for the sake of clarity (solid curve). Right-hand ordinate axis—absorption coefficient at low irradiation intensity. The lower horizontal scale indicates the CO$_2$-laser emission lines.

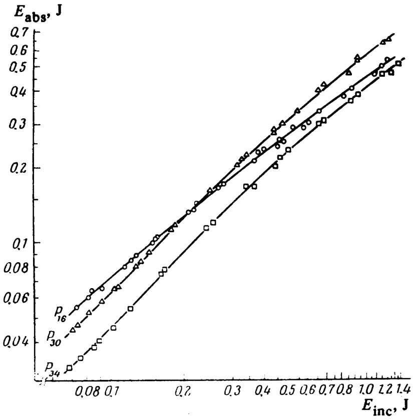

FIG. 4. Dependence of the absorbed energy on the incident energy at various CO$_2$-laser frequencies (SF$_6$ pressure 2.7 Torr). Solid curves—result of statistical reduction of several measurement runs; ○, △, □—experimental points of single run.

The variation of the energy absorbed in the cell with changing incident-power density (at the cell entrance), in the range from 1.5×10^4 to 10^7 W/cm^2, was investigated at fixed incident radiation wavelength (the irradiating-pulse duration at half height was ~ 3 μsec, and the irradiated area was 0.3 cm^2). The most interesting range, from our point of view, of variation of the energy incident on the cell (intensities (1–4×10^5 W/cm^2) is shown in Fig. 4. It follows from the presented data that in this band the maximum of the absorption (of the resonant frequency) is shifted towards the long-frequency region, thus indicating the presence of strong energy-transfer processes in the case of vibrational excitation of the molecules. The same figure shows data on the absorption of IR radiation at a frequency 930 cm^{-1} by SF$_6$ molecules (P_{34} line of the CO$_2$ laser). It is seen that in the entire range of the investigated irradiation intensities the absorption at this frequency is much lower, even though this frequency corresponds to the maximum of the linear absorption of ^{34}SF$_6$.[4]

We measured the IR energy passing through the cell at various pressures of the absorbing gas. These results are shown in Fig. 5a. The reduction of these data shows that the energy absorbed in the mixture depends on the SF$_6$ pressure in a nonlinear manner. Addition of hydrogen to the SF$_2$ (in the ratio SF$_6$: H$_2$ = 2.5 : 1) leads to a change in the absorbed energy at both IR frequencies. From the obtained data we determined the average number of CO$_2$-laser quanta absorbed by one SF$_6$ molecule:

$$n = \frac{E_{abs}}{N},$$

where N is the total number of absorbing molecules in

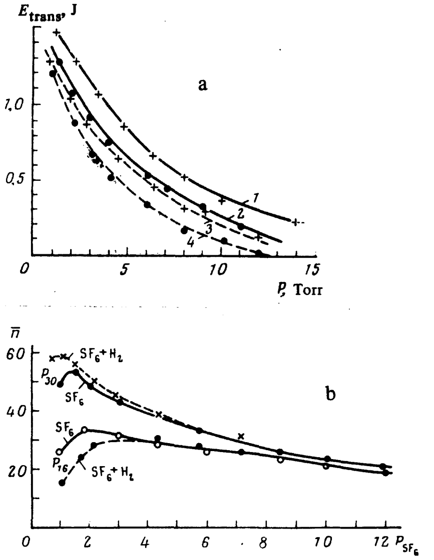

FIG. 5. a) Dependence of the energy passing through the cell on the gas pressure at two incident radiation from the CO_2 laser: 1, 3—mixture SF_6 : H_2 = 2.5 : 1; 2, 4—SF_6. 1, 2—transition P_{16}; 3, 4—transition P_{30}. b) Dependence of the average number of absorbed quanta of two CO_2-laser frequencies, per SF_6 molecule, on the pressure of the absorbing gas. In both cases (a and b) the irradiation intensity at the entrance to the cell was ~ 2×10^6 W/cm^2.

the irradiated volume. It is seen from the results (Fig. 5b) that at a fixed level of the intensity incident on the cell (~ 2×10^6 W/cm^2) the value of n reaches its maximum at a certain SF_6 pressure. Dilution of the SF_6 with hydrogen changes the character of the absorption of the CO_2-laser radiation. Addition of the hydrogen increases the fraction of the absorbed energy when the mixture is exposed to long-wave quanta, and when shorter wavelength radiation acts on the mixture the absorption decreases. A gradual decrease of n is observed with increasing SF_6 pressure.

3. THE CHEMICAL REACTION SF_6+H_2 INITIATED BY CO_2 RADIATION

It is of interest to trace how the singularities in the absorption of high-power IR laser radiation by the molecules manifest themselves in the initiation of chemical reactions. The subject of the investigation was chosen to be the chemical reaction SF_6+H_2. We investigated the induced radiation of excited HF* molecules, which are one of the products of the reaction of vibrationally excited SF_6 molecules with hydrogen. The energy and time characteristics, as well as the spectral composition of the radiation of this chemical laser have been investigated in sufficient detail before.[6-8] In the cited studies, the frequency of the initiating radiation of the CO_2 laser was chosen such as to maximize the SF_6 absorption.

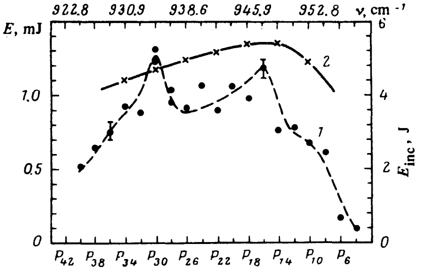

FIG. 6. 1) Generation energy of HF chemical laser (left-hand ordinate axis) initiated by CO_2-laser radiation of varying frequency (concentration SF_6 : H_2 = 2.5 : 1, pressure 4 Torr). 2) Distribution of the energy incident on the cell (right-hand ordinate axis). The lower horizontal line indicates the CO_2-laser emission lines.

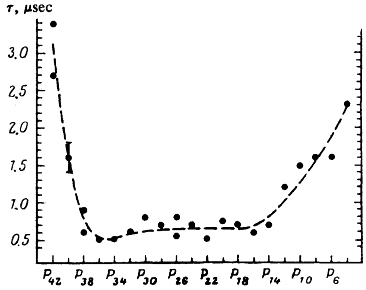

FIG. 7. Delay of the HF lasing pulse vs the incident-radiation frequency (the experimental conditions are indicated in Fig. 6). The lower horizontal line shows the CO_2-laser emission lines.

We investigated the HF lasing energy at various frequencies of the initiating pulse (Fig. 6). The working medium of the HF laser was the mixture SF_6+H_2 (2.5 : 1) with a total pressure 4 Torr, corresponding to the maximum of the HF-lasing energy yield.[8] It follows from the presented data that at an incident-radiation density (5–7) · 10^6 W/cm^2, two maxima of the HF-lasing energy yield are observed, corresponding to the P_{30} and P_{16} transitions of the CO_2 laser (Fig. 6, curve 1). With decreasing intensity of the initiating pulse, these maxima become "smeared out." The threshold values of the radiation incident on the cell, at which the HF lasing stops, lies in the range (1.5–7) · 10^5 W/cm^2 for different lines of the CO_2 laser.

The measured values of the delay of the instant when the HF lasing pulse is produced following the start of the initiating pulse are shown in Fig. 7. The relative distribution of the intensity over the spectral lines of the CO_2 laser is shown by curve 2 of Fig. 6. It is seen that at incident radiation intensities (4–7) · 10^6 W/cm^2, in the frequency band corresponding to the transitions $P_{16}, \ldots, P_{36}$ of the CO_2 molecule, this delay is minimal and remains practically constant. With decreasing irradiating-pulse intensity, and also when the applied IR frequency lies at the edges of the absorption band of the SF_6 molecule, an appreciable increase of the delay is observed, reaching 2.0–3.0 μsec.

It must be noted that reproducibility of the data shown in Figs. 6 and 7 is attained only if the CO_2 laser radiation falls exactly in the caustic of the HF-laser resonator. Violation of this condition leads to a decrease in the energy of the HF lasing and to an increase in the delay between the starts of the CO_2 and HF pulses.

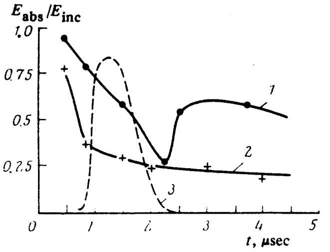

FIG. 8. Change in the fraction of the absorbed energy during the time of the irradiating pulse at two CO_2-laser frequencies: 1—$\nu \approx 934$ cm^{-1}; 2—$\nu \approx 948$ cm^{-1}. The dashed curve (3) shows the shape of the HF lasing pulse (experimental conditions of Fig. 6).

We have investigated the time dependence of the absorption of the CO_2-laser pulse at various wavelengths. Figure 8 shows the time dependence of the ratio of the energy absorbed in the cell to the incident energy at two frequencies (the transitions P_{16} and P_{30} of the CO_2 molecule). In both cases, a maximum of the absorption is observed 0.5 μsec after the start of the irradiating pulse. As follows from the experimental results, when the radiation at the frequency $\nu = 934$ cm^{-1} is absorbed, the medium experiences a noticeable bleaching, the maximum of which coincides with the end of the HF-lasing pulse (2.0–2.5 μsec after the start of the initiation).

To increase the effectiveness of the excitation of the SF_6 molecules, and consequently to increase the energy yield of the HF laser, the $SF_6 + H_2$ mixture was exposed to IR at two wavelengths. The transverse dimensions of the active region of the employed CO_2 laser have made it possible to place in the resonator two diffraction gratings at the same time. Measures were taken to prevent interaction of the two resonators via the mutual participation of higher-order transverse modes in the lasing. The same cell volume was simultaneously irradiated by both frequencies with the aid of an additional mirror. The two diffraction gratings ensured separation of individual CO_2 laser lines in the ν_3 absorption band of the SF_6 molecule. The same experiments have revealed an increase in the efficiency of the conversion of the CO_2 laser energy into HF generation. The maximum increase in the efficiency of conversion, by a factor 1.4, was obtained when the $SF_6 + H_2$ reaction was initiated by radiation at two wavelengths of equal intensity, corresponding to the transitions P_{16} and P_{30} of the CO_2 molecule.

4. DISCUSSION

The shift of the maximum of the ν_3 absorption band towards the red shows that when a molecule possessing anharmonicity is excited, an appreciable contribution to the absorption can be made in the saturation regime by transitions between excited vibrational levels. The number of resonantly excited levels depends on the width of the emission line and on the anharmonicity of the molecule vibrations. The decrease of the transition frequency with increasing number of the vibrational levels, due to the anharmonicity, can be offset by increasing the frequency in the rotational transitions. This circumstance has already been pointed out earlier.[9,10] In order for the maximum number of successive rotational-vibrational transitions to fall into resonance, it is necessary to choose the radiation frequency as low as possible. The first should be a P-branch transition with a large rotational quantum number J. In order to compensate for the anharmonicity, the subsequent rotational-vibrational transitions must begin with a level having a smaller value of J (if this is a P transition), or alternate successively with the Q and R transitions. Choosing the laser-radiation frequency

$$\nu_n = \nu_{10} - 2BJ_k, \tag{1}$$

it is possible to realize almost resonant population of the $v = 2k + 1$ level if

$$J_k = {}^1/_2 k \Delta\nu_a / B, \tag{2}$$

where D is the rotational constant of the molecule; J_k is the angular momentum quantum number of the molecule; ν_{10} is the frequency of the vibrational transition $v = 0 \rightarrow v = 1$; $\Delta\nu_a$ is the anharmonicity factor.

When rotational equilibrium is established in the system (in our case $\tau_p \sim 10^{-16}$ sec $\gg \tau_{r-r}$), each successive transition to a higher vibrational state can begin with any level J. It might seem that the larger J for the initial transition, the lower the initial resonant frequency of the laser, meaning that a transition of the molecule to a higher vibrational state is possible. However, the use of transitions with very large J is difficult: the intensity of the rotational-vibrational transitions (the effect of cross section of the transition) decreases sharply with increasing J, like

$$J^2 \exp[-BJ(J+1)/kT] = J^2 \exp(-J^2/J_m^2).$$

For SF_6 we have $J_m \approx 50$. The level scheme in Fig. 9 explains the possible mechanism whereby the CO_2-laser radiation is absorbed by the SF_6 molecule.

We present numerical estimates concerning our experiments. The frequency of the P transition $v = 0$, $J \rightarrow v = 1$, $J - 1$ is given by the relation[11]

$$\nu_{10} = \nu_3 - 2B(1-\zeta)J.$$

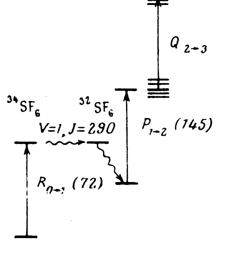

FIG. 9. Cancellation of the anharmonic frequency shift by the rotational energy of the molecule.

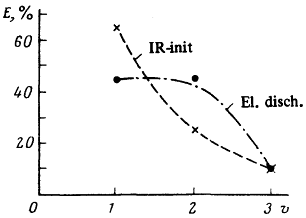

FIG. 10. Distribution of the HF-radiation energy (over the vibrational levels—v) for two methods of initiation of the chemical reaction SF_6+H_2; on the abscissa axis are shown the vibrational quantum numbers of the upper level.

According to the experimental and theoretical data,[5,11,12] $\nu_3=948.2$ cm^{-1}; $B\approx 0.09$ cm^{-1}; $\zeta=0.73$. The anharmonicity factor for the SF_6 molecule amounts to $\Delta\nu_a\approx 6-7$ cm^{-1}.[13] A shift of the laser frequency by 14 cm^{-1} relative to ν_0 towards lower frequencies (see Fig. 3) makes it possible to realize the following cascade of resonant transitions: $P_{0\to 1}$ (14 cm^{-1}); $P_{1\to 2}$ (−7 cm^{-1}); $Q_{2\to 3}$; $R_{3\to 4}$ (+7 cm^{-1}); $R_{4\to 5}$ (+14 cm^{-1}) etc. However, if the absorption of the frequency (ν_0-14) cm^{-1} is attributed to $P_{0\to 1}$ in $^{32}SF_6$, then this transition would correspond to $J=290$. The intensity of P (290) is 10^{-12} of the most intense transition $(P_{0\to 1}(J_m))$, thus indicating the impossibility of the first step in this cascade on account of the $P_{0\to 1}$ (290) transition. However, our mixture contains ~4.4% of the isotopic modification of the molecule $^{34}SF_6$. The frequency 934 cm^{-1} corresponds to the $R_{0\to 1}$ (72) transition of this molecule, i.e., it falls practically in the region of the maximum of absorption of $^{34}SF_6$. We can therefore propose as the first step in the absorption the $R_{0\to 1}$ (72) transition in $^{34}SF_6$, followed by rapid transfer of the excitation of the $^{32}SF_6$ molecules: $P_{1\to 2}$ (145); $Q_{2\to 3}$, $R_{3\to 4}$ (145). The intensities of the P and R transitions with $J=145$ are smaller by only a factor of 200 than the maximum intensity (with $J_m=50$), this indicating the feasibility of this cascade of successive transitions.

The proposed mechanism is valid at high intensities of the incident radiation ($I>3.9\cdot 10^5$ W/cm^2). Lowering the intensity shifts the maximum of the absorption towards shorter wavelengths, and at $I<10^5$ W/cm^2 the absorption maximum occurs at the frequency $\nu=948$ cm^{-1}, corresponding to the usual absorption of the SF_6 molecules.[5] In the saturation regime, the absorbed energy E_n can be estimated in the following manner. In a cascade of end transitions we have $E_n=h\nu nN_0/2$, where N_0 is the total number of the lower-level molecules participating in the absorption. In our case $n=4$, so that $E_1=2h\nu N_0$. At the frequency of the P_{16} transition of a CO_2 laser, a cascade of two transitions is possible: $0\to 1$, to which a contribution can be made by the transitions Q: R (J_m) and P (J_m), and the transition $R_{1\to 2}$ (145). Therefore the absorbed energy at the frequency 948 cm^{-1} should not exceed $E_2=h\nu N_0$, which is half the expected absorption at the frequency 934 cm^{-1}. Experiment (see Fig. 3) shows that $E(934\text{ cm}^{-1})/E(948\text{ cm}^{-1})=1.6$, in satisfactory agreement with the approximate theoretical prediction.

It is observed in experiment (see Fig. 6) that the HF-laser energy yield, if the reaction is initiated by radiation having an intensity ~6×10^6 W/cm^2 at a frequency $\nu=934$ cm^{-1}, is higher: $E(P_{30})/E(P_{16})=1.25$. Since the reaction SF_6+H_2 is not exothermal (it proceeds only by virtue of the absorbed energy), this fact is easily explained on the basis of the results of experiments on absorption (see Fig. 5b).

In the case of IR initiation, the molecules most likely to react with the hydrogen are vibrationally-excited SF_6, whereas in electric initiation of this reaction gives rise to separation of atomic fluorine, which reacts with H_2. This is evidenced by the sharp change in the generation energy spectrum on going from one method of initiating the reaction to the other[3] (Fig. 10).

5. CONCLUSION

It can be noted in the presented results that the singularities observed by us in the experiments on resonant absorption of laser IR radiation take place at high irradiation intensities and at large pulse durations, ~3 μsec. On the contrary, it was observed in[1] that the most effective frequency for the separation of the sulfur isotope ^{32}S is shifted by 7 cm^{-1} away from the resonant mode towards larger wavelengths. This result is attributed to the existence of a mechanism for collisionless excitation of the molecules in the absence of relaxation over the rotational levels during the irradiation time (the duration of the irradiation pulse is shorter than the times of vibrational and rotational relaxation). In these experiments, the radiation interacts with a small fraction of the total number of the molecules in the irradiated volume. The frequency dependence of the absorbed energy under these conditions correlates with the shape of the linear absorption band, since the absorption coefficient at a given frequency is determined principally by how many molecules on individual rotational levels interact with the radiation.[14] In contrast, a pulse with duration τ_p longer than the time τ_{r-r} of the rotational relaxation is capable of involving in the excitation process practically all the molecules that are distributed over the rotational levels. However, in order to prevent loss of selectivity of the process, the rate of rotational relaxation must exceed the rate of energy exchange between the different isotopes. If the rate of the chemical reaction, which leads to the enrichment of the reaction products by the required isotope, exceeds in this case the time of the $v-T$ relaxation, then this isotope-separation process can turn out to be quite effective. To this end, the gas of the exciting molecules should be strongly diluted by a reagent that quenches relatively slowly all the vibrationally excited molecules, but entering rapidly in a chemical reaction with these molecules and causing their rapid relaxation over the rotational levels. We note that in our experiments the addition of a non-absorbing gas to SF_6 led to different changes in the values of the energy absorbed at different frequencies. This circumstance must be taken into account when a gas of resonantly absorbing molecules is strongly diluted.

We have performed trial experiments on the separation of sulfur isotopes in the reaction of the molecule

SF_6 with H_2S. When the $SF_6 + H_2S$ mixture was exposed to CO_2 laser radiation of frequency $\nu = 948$ cm^{-1}, a shift of the isotopic ratio of the residual gas towards the ^{34}S isotope was attained.

It was shown recently[15] that $v-T$ relaxation of CO dissolved in liquid nitrogen is strongly suppressed. It is possible that this is also true of other molecules (for example, solutions of SF_6 in liquid oxygen or nitrogen). These solutions should satisfy then all the above-mentioned requirements needed to ensure selectivity of the process when the molecules are excited with a "long" pulse of laser radiation ($\tau_p > \tau_{r-r}$), and could be used successfully for isotope separation and for other laser-chemical processes. Finally, we note that at low temperature the process can be made selective even at an appreciable energy exchange between the isotopes.[16]

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Observation of radiofrequency coherence during the excitation of hyperfine structure transitions in optically oriented atoms

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Radiofrequency coherence of states corresponding to the Zeeman sublevels of optically oriented cesium atoms during the excitation of hyperfine structure transitions by amplitude-modulated microwave magnetic field is reported. The coherence signals were recorded both by varying the modulation frequency and by varying the frequency of the microwave field. The signals have a different shape and behavior, depending on the intensity of the microwave magnetic field. The observed phenomena are examined theoretically, and the theoretical results are compared with experimental data.

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Experiments on the optical orientation of atoms have revealed a modulation of light absorption both at the transition frequency between the Zeeman sublevels of one of the hyperfine structure levels[1] and at the transition frequency between different hyperfine structure levels.[2] The light intensity is then found to be modulated at the frequency of the magnetic resonance. Light modulation occurs because the magnetic resonance is accompanied by the coherent superposition of states by the optically oriented atoms, and this is manifested microscopically by the appearance of a component of the resultant magnetic moment which precesses around the constant magnetic field, or oscillates with the resonance frequency, and gives rise to the modulation of absorp-

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