Excitation of drift motion of multiatomic molecules by resonance IR radiation

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The results of an experimental study of the drift motion of SF_6 molecules under the influence of CO_2 laser radiation are reported for the first time. The concentration ratio of SF_6 molecules (in mixtures with He and H_2) at the input and output ends of the cell reached $\sim 10^2$. The fluxes of resonance molecules exceeded several billion fold those produced in the experiments[3,8] with sodium vapor as a result of irradiation by optical range sources, although the average radiation power directed into the cell did not exceed ~ 20 W.

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As shown in Ref. 1, internal macroscopic fluxes due to the drift motion of resonance molecules can arise under the action of IR radiation in a gas mixture. This effect is related to the one examined in Refs. 2 and 3 for atomic gas. The use of lasers in the IR range and molecular gases has a number of advantages (see Ref. 1). First, there are radiation sources in the IR range which greatly exceed in average power and efficiency the lasers in the visible and UV range (for example, CO_2 lasers). At the same time, the energy cost of the process for irradiation with a IR radiation source is smaller by a factor of $(M/m)^{1/2}$ than that in the optical range (M is the mass of a molecule or an atom and m is the electron mass). Second, the complex vibrational-rotational spectrum of multiatomic molecules reduces considerably the requirement for the line width of generation and frequency tuning of the source. Finally, the lower operating temperatures (than those for atomic gas) and much larger densities facilitate the implementation of the effect (gas scrubbing, ecology, isotope separation, etc.).

In irradiating the molecular gas by a IR laser, in addition to the drift effect there can also arise the effects of diffusive suction of resonance particles into a beam[4] and of thermal diffusion stimulated by the laser action.[5] Both effects, which have a diffusive nature, can be eliminated by using a pulse-periodic radiation with a large off-duty factor $N = (f\tau_\rho)^{-1}$ (f is the repetition frequency and τ_ρ is the pulse duration). During the pauses between the pulses the ordinary diffusion dissipates the fluxes produced during laser irradiation.

The experiment was performed by using a pulse-periodic CO_2 laser described in Ref. 6. The SF_6 molecules at a pressure of 0.1 +1 Torr in a mixture of buffer gas (He or H_2 at a pressure in the range of 0 to 30 Torr) were used as resonance particles. The CO_2 laser radiation with a pulse duration $\tau \approx 5$ μsec, a variable pulse energy (0.1 +1 J), and a pulse repetition frequency (up to 200 Hz) was directed into a 1-m-long copper cell with a 1×3-cm^2 cross section. The average power at the entrance to the cell was ~ 20 V.

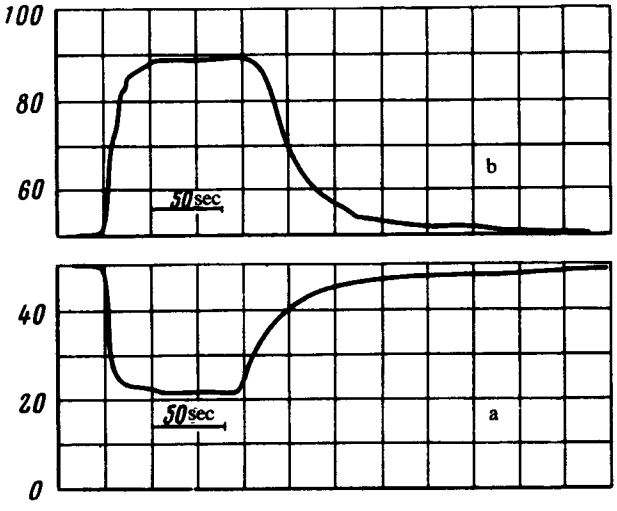

FIG. 1. Readings of the IKS-29 spectrophotometer (a) at the input end (relative to the beam) and (b) at the output end of the cell. SF_6:He = 1:10, $P_\Sigma = 4$ Torr, $f = 50$ Hz.

The ends of the cell had 10-cm-long extensions through which the SF_6 density was continuously recorded by means of the IKS-29 spectrophotometer. The gas samples could be taken from both ends of the cell and the IR and mass spectra could be recorded before and after irradiation. The diffraction grating made it possible to tune the CO_2 laser radiation along the rotational lines in the 10.6-μm band.

Figure 1 shows the readings of the IKS spectrophotometer at the input end (rela-

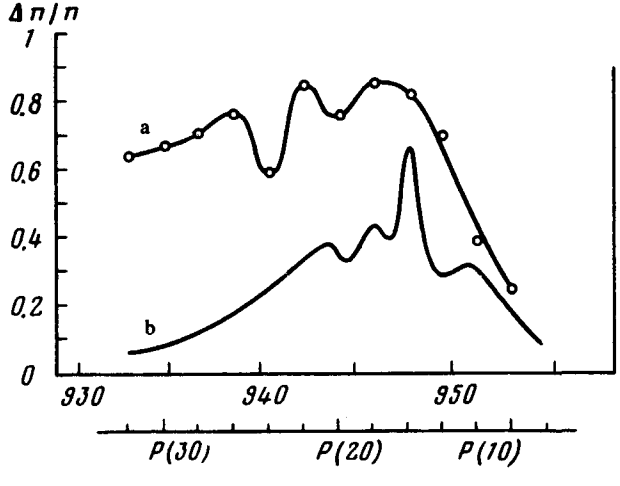

FIG. 2. (a) Dependence $(\Delta n/n)_{SF_6}$ at the output end of the cell on the wavelength of the CO_2 laser; (b) absorption spectrum of SF_6, SF_6:He = 1:11, $P_\Sigma = 4$ Torr, $f = 80$ Hz.

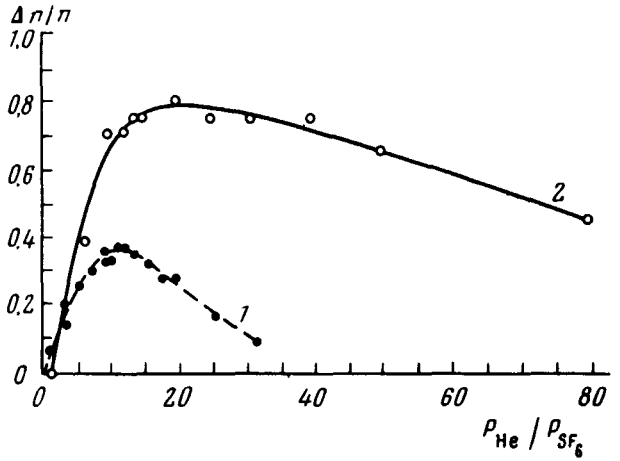

FIG. 3. Dependence of $(\Delta n/n)_{SF_6}$ at the output end of the cell on the ratio $p(He)/p(SF_6)$: curve 1, $P_{SF_6} = 0.5$ Torr; curve 2, $P_{SF_6} = 0.2$ Torr.

tive to the beam) and the output end of the cell. An increase of SF_6 concentration can be seen at the dependence of the change in density of the SF_6 molecules (Δn) on the following factors: the energy in a single CO_2 laser pulse, pulse repetition frequency, total pressure of the mixture, relative content of $n(SF_6)/n(He)$, and the wavelength of exciting radiation. Figure 2 shows the spectral dependence of $(\Delta n/n)_{SF_6}$ at the output end of the cell. In the investigated frequency range the sign of the effect remains the same and there is a red frequency shift of the maximum relative to the linear absorption spectrum of $^{32}F_6$, which apparently means that molecules excited to the upper vibrational states 1, 2, etc. are drawn into the drift. Figure 3 shows the dependence of $\Delta n/n$ at the output end of the cell on the ratio $p(He)/p(SF_6)$. As the repetition frequency f increases, $(\Delta n/n)_{output}$ increases monotonically and approaches unity. A decrease in density at the $\sim 10\%$ level at the output end of the cell can be observed even at a repetition frequency of $f \sim 1$ Hz, which indicates that the observed effect has a nondiffusive nature. The value of $(\Delta n/n)_{output}$ increases with increasing energy in the radiation pulse. There was no evidence of a change in density when SF_6 was irradiated in the absence of a buffer gas. In the dependence of the mixture on the total presure P_Σ for the ratio SF_6: He = 1:11) we observed a maximum of $(\Delta n/n)_{output}$ at a pressure of $P_\Sigma \sim 3$–4 Torr (in different lines of CO_2 radiation).

Analogous dependences can be observed if hydrogen is used as a buffer gas. The maximum value of the effect n_{input}/n_{output} amounts to $\sim 10^2$. A noticeable variation of the effect was not observed when an external, stationary, low-pressure CO_2 laser[7] was used to narrow down the source lines (here $\Delta\nu_l \sim 20$ MHz).

We performed experiments by irradiating a 1-m-long glass cell with a 5-mm inside diameter using a stationary CO_2 laser which is line tunable with the help of a diffraction grating. At the power level $P \sim 30$ W/cm^2 the variations of SF_6 density (in a mixture with He at P_Σ to 30 Torr, $p_{SF_6} \geqslant 0.1$ Torr) at both ends of the cell did not exceed the measurement errors.

The preliminary experiments on irradiation of the mixture $^{32}SF_6$:$^{34}SF_6$ = 1:3.7 (at He pressure from 0 to 30 Torr) showed a $^{34}SF_6$ enrichment at the input end of the cell and a $^{32}SF_6$ enrichment at the output end. The total enrichment, determined by the relation

$$\frac{n(^{34}SF_6)}{n(^{32}SF_6)}\bigg|_{in} = \frac{n(^{34}SF_6)}{n(^{32}SF_6)}\bigg|_{out},$$

amounts to 1.08 ± 0.03.

As mentioned in Ref. 1, the nonresonance gases can be separated by means of the drift of resonance molecules, since their velocities are generally different.

In the mixture with SF_6:H_2:He (0.3 : 1.8 : 1.8; $P_\Sigma = 4$ Torr) we observed an increase in the hydrogen content relative to He content in the samples selected for the mass-spectral analysis at the output end of the cell, at the $\sim 20\%$ level.

The relations for characteristic stabilization time (τ_s), for the densities of molecules (n_{input}/n_{output}), and for the decay time (τ_d) after the laser is turned off favor the drift nature of the observed effect.

Let us examine the diffusion problem with allowance for drift deflection of particles with the average velocity $\langle u \rangle = u_{max}/N$; u_{max} is the maximum drift velocity; in the pulse-periodic case ($\tau_\rho < \tau_{quench}$) $\mu_{max} \sim (\Delta\nu/\nu)[\nu_B/(\nu_B + \nu_s)]l/\tau_\rho$; here $\Delta\nu$ is the difference in the transport frequencies of molecules in the upper and lower states, ν_B and ν_S are the frequencies of collision of molecules with the buffer and with each other, l is the path length relative to the momentum transfer $l \sim v_T/(\nu_B + \nu_S)$. At $n \ll n_B$ we can see that during the time $\tau_s = (2L/\langle u \rangle)[(2\pi/P_e)^2 + 1]^{-1}$ (here $P_e = \langle u \rangle L/D$ is the Peclet number) the distribution of the resonance particles is established, where $n_{input}/n_{output} = e^{P_e}$. This distribution decays in the time $\tau_d = L^2/\pi^2 D$ after the laser is turned off. Using the experimental values $P_e = 1 - 4$, $\tau_s \sim 5$ sec, and $\tau_d \sim 25$ sec, we obtain $\langle u \rangle \sim 1 + 10$ cm/sec. Taking into account the off-duty factor $N \geqslant 10^3$, we have for the maximum velocity $u_{max} \sim 10 - 10^2$ m/sec. It follows from the expression for u_{max} that the optimum value of the effect is reached at $\nu_B \sim \nu_S$. Taking into account the relations $\nu_i \sim [M/\mu_i]^{-1} n_i(\sigma n)_i$ [where μ_i is the reduced mass of a resonance particle and of its partner (i) and n_i is its concentration], we obtain $(n_B/n_S)_{opt} \sim (\mu_s/\mu_B)^{1/2}(\sigma_s/\sigma_B) \sim 12$, in good agreement with the experiment (Fig. 3).

It follows from the given data that the fluxes of molecules attained under our conditions exceeds several billion fold those of the atoms in the experiments.[3,8] This excess is due to a larger density of resonance particles, shorter steady-state establishment time, and a larger cross section of the cell.

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[1]A. M. Dykhne and A. N. Starostin, Dokl. Akad. Nauk SSSR **251**, 1 (1980). [Sov. Phys. Dokl., (1980) (in press)].
[2]F. Kh. Gel'mukhanov and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. **29**, 773 (1979) [JETP Lett. **29**, 711 (1979)].
[3]V. D. Antsigin, S. N. Atutov, F. Kh. Gel'mukhanov, G. G. Telegin, and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 262 (1979) [JETP Lett. **30**, 243 (1979)].

[4]F. Kh. Gel'mukhanov and A. M. Shalagin, Zh. Eksp. Teor. Fiz. **77**, 461 (1979) [Sov. Phys. JETP **50**, 234 (1979)].
[5]N. V. Karlov, Yu. N. Petrov, A. M. Prokhorov, and I. V. Fedorov, Zh. Tekh. Fiz. **48**, 2334 (1978). [Sov. Phys. Technical Phys. **23**, 1337 (1978)].
[6]V. Yu. Baranov, E. P. Velikhov, S. A. Kazakov, D. D. Malyuta, V. S. Mazhevov, V. G. Niz'ev, S. V. Pigul'skiĭ, V. D. Pis'mennyĭ, and A. I. Starodubtsev, Kvantovaya Élektronika **6**, 811 (1979) [Sov. J. Quant. Electronics **9**, 480 (1979)].
[7]V. Yu. Baranov, S. A. Kazakov, V. V. Likhanaskiĭ, V. S. Mezhevov, A. P. Napartovich, M. Yu. Orlov, V. D. Pis'mennyĭ, and A. I. Starodubtsev, Kvantovaya Elektronika **6**, 2463 (1979) [Sov. J. Quantum Electron. **9**, 1456 (1979)].
[8]J. E. Bjorkholm, A. Ashkin, and D. B. Pearson, Appl. Phys. Lett. **27**, 534 (1975).