Diffusion of SF_6 under the action of CO_2 laser radiation[1)]

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The diffusion of SF_6 gas in a mixture with He buffer gas under the action of CO_2 laser radiation is investigated in this work as a function of the detuning of the laser frequency relative to the center of the absorption line of SF_6. Within the limits of accuracy of the experiment, the change in the gas density did not depend on the sign of the frequency detuning. The process observed is explained by laser-driven thermal diffusion and is not related to light-induced drift.

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Under the action of continuous IR radiation, absorbed resonantly by molecular gases in the pressure range 0.1–10 Torr, there is a change in the partial concentrations of the components of the mixture. In Refs. 1 and 2, this was related to laser thermodiffusion processes. In Refs. 3–5, the separation of the mixtures was related to the appearance of light-induced drift (LID).[6,8] Thus, effects occurring under similar experimental conditions and having similar manifestations were interpreted as different physical phenomena and this problem still has not been clarified. Direct confirmation of the existence of light-induced drift should be the dependence of the diffusion processes in molecular gases, irradiated by laser radiation, on the sign of the detuning of the exciting laser frequency relative to the center of the transition of the absorbing gas. In experiments on light-induced drift described in the literature,[3–5] such detuning was not realized. Starting from this fact, we set up an experiment on the observation of light-induced drift in the gas mixture SF_6 + He under conditions such that the frequency of the exciting laser was detuned relative to the center of the absorption line of SF_6.

The investigations were carried out in copper and glass cells. The inner diameter of the copper cell was 4 mm and that of the glass cell was 4 mm. The length of both cells was 18 cm. The side extensions for detecting changes in absorption in the mixture studied, which were situated on both end faces of the copper cell, had a thickness of 4 mm and allowed carrying out absorption measurements near the axis of the cell (zone 1) and at distances up to 10 mm from it (zones 2 and 3)

Two CO_2 lasers were used in the experiments (Fig. 1). The first was used to create the exciting field in the cell and the second was used to create the probing field for detecting absorption in the side extensions of the cell. The frequency of the probing laser radiation [the $P(18)$ line of the transition 00°1–10°0 in CO_2 in the single-mode regime] was stabilized by the resonance saturation of absorption of SF_6 in the entire outer cell to within ~100 kHz. The powers of the exciting and probing lasers were 5 W and 100 mW, respectively. The relative change in the SF_6 concentration in different zones of the cell was detected by the change in the intensity of the probing radiation at the outlet from the cell to within 0.1–0.3%. The gas pressure in the cell was varied in the range 0.3–1 Torr for SF_6 and 2–10 Torr for He.

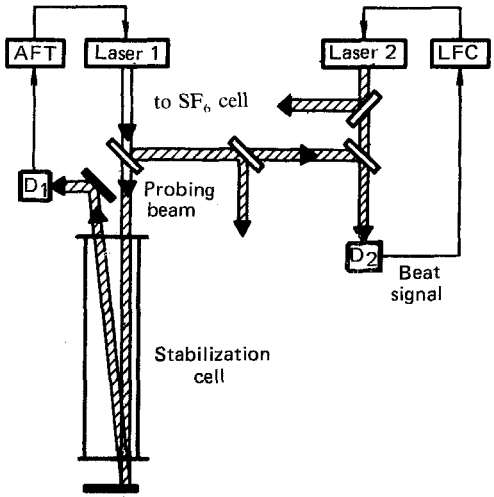

FIG. 1. D_1 and D_2 are photodetectors, AFT is the automatic frequency tuning system, LFC is the system coupling the frequency of laser 2 to the frequency of the stabilized laser 1 ($\Delta = \pm 10$ MHz).

Probing was carried out in three zones of the cell: 1) hot, and 2) and 3) cold (Fig. 2). In zones 2 and 3, the laser, creating the exciting field, as tuned to the $P(18)$ line and in zone 1 the measurements were performed for the three lines $P(18)$, $P(20)$, and $P(28)$. For measurements on the $P(18)$ line, the frequency of the exciting laser was detuned relative to the absorption line of SF_6 by $+10$ or -10 MHz.

In all cases, we observe two types of changes in the probing radiation intensity: fast (of the order of several ms) and slow (of the order of several minutes), similar to that described in Ref. 5. The fast changes, occurring at the time the exciting field was switched on and off, are attributable to thermal effects, leading to a change in temperature and gas density.

The investigations, carried out in zones 1, 2, and 3 in switching on the exciting field, showed that the fast processes, related to thermal effects, have the same sign in zones 2 and 3 (increase in SF_6 concentration), while in zone 1 they had the opposite

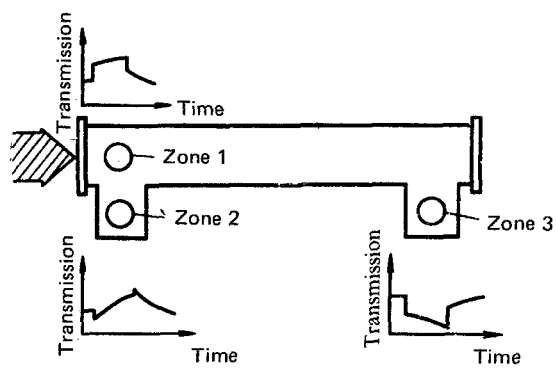

FIG. 2. Absorbing cell.

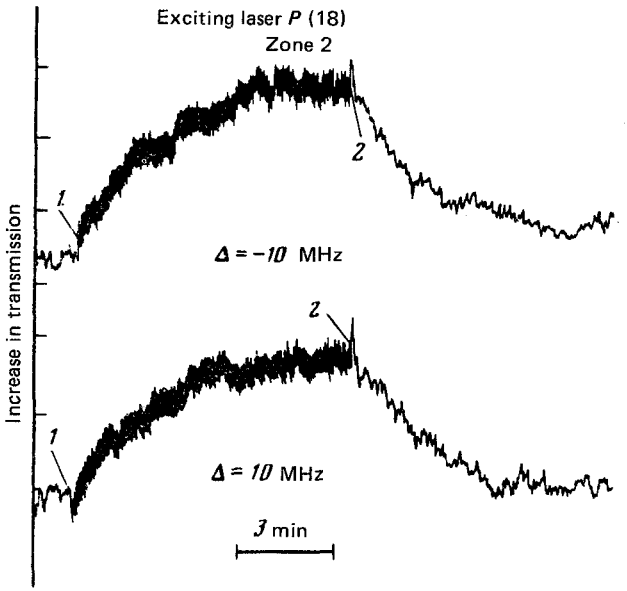

FIG. 3. The change in transmission of the probing radiation in zone 2. The radiation of the exciting laser is detuned by -10 MHz for the upper curve and by $+10$ MHz for the lower curve from the center of the $P(33)$, A_2^1 of the ν_3 band, absorption line of SF_6. 1) Time at which the exciting laser was switched on; 2) time at which the exciting laser was switched off.

sign. We are interested in the processes responsible for the slow changes. The latter have the same sign and correspond to a decrease in concentration in zones 1 and 2 and an increase in concentration in zone 3, when zone 3 is the cold zone. With a decrease in the coefficient of absorption, when the radiation of the exciting laser was not completely absorbed, the change in the concentration in zone 3 changed sign. This sign change can be observed with detuning on other lines, when the absorption coefficients can differ greatly. In Ref. 5, this effect was interpreted as light-induced drift. Typical traces of the processes occurring are presented in Fig. 3, which show the results of measurements in zone 2. Points 1 and 2 correspond to switching on the exciting radiation in the cell. The exciting radiation was switched slowly ($\sim$0.1–0.3 s) and, therefore, the effects of barodiffusion were eliminated, while the results of experiments in Refs. 3 and 9 are explained by barodiffusion.

The change in sign of the detuning of the exciting laser frequency from the center of the SF_6 line (Fig. 3) does not change the nature or magnitude of the change in concentration. In this case, a 10% change in the power of the exciting laser with frequency detuning was taken into account. Since neither the magnitude nor sign of the effect depends on the frequency detuning, the separation processes are naturally explained by laser thermodiffusion.

Light-induced drift is based on the differences between the collision frequencies (or cross sections) in the ground and excited states. Using the expression[7] for the relative change in concentration

$$\frac{\Delta N}{N} = \frac{\nu_n - \nu_m}{\nu_m} \frac{2\Delta S}{Nh\omega\bar{v}} \phi(\Omega),$$

where $\nu_n - \nu_m/\nu_m$ is the relative difference in the collision frequencies for the upper and lower levels, ΔS is the change in the energy flux density on passage through the cell, $\bar{v}$ is the average thermal velocity, $\phi(\Omega)$ is a factor that depends on the quantity Γ/kv and the detuning of the laser frequency from the center of the absorption line, it is possible to estimate the relative difference between the collision frequencies in the upper and lower levels, which corresponds to the limiting sensitivity of the experiment. The highest sensitivity for observing light-induced drift under conditions of our experiment could be attained for SF_6 pressures ~ 0.3 Torr and He pressures ~ 2 Torr. The absorbed power in this case was 3 W. We chose the factor $\phi(\Omega)$ as equal to ~ 0.25. Under these conditions, we found that $(\nu_n - \nu_m)/\nu_m = 3\times 10^{-4} - 6\times 10^{4}$. The difference between the collision frequencies in the upper and lower levels can be estimated from spectroscopic data in Ref. 10, where the SF_6 line broadening constants were measured in unidirectional and oppositely traveling waves and constituted 4 ± 0.2 and 3.8 ± 0.3 MHz/Torr, respectively. We can conclude from this that the collision frequencies in the upper and lower levels differ by an amount $\sim 5\times 10^{-2}$. Certain results concerning the difference in the collision cross sections can be obtained from data on the line shift. Experimental estimates on shifts of SF_6 lines as a function of concentration give the quantity 70–200 kHz/Torr. It can be shown that the cross sections in this case must differ by an amount 10^{-2}. Apparently, the absence of light-induced drift is explained by other reasons and not by the small difference between the collision frequencies in the ground and excited states.

Thus, the experimental results have shown that the basic process responsible for separation of components of a gas mixture consisting of SF_6 and He under the action of continuous laser radiation is the process of laser thermodiffusion.[1,2] No effect of light-induced drift on the change in the concentration of gases within the limits of accuracy of our experiment was observed.

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