

# Separation of sulfur isotopes with enrichment coefficient $> 10^3$ through action of $\text{CO}_2$ laser radiation on $\text{SF}_6$ molecules

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We report separation of sulfur isotopes by selective dissociation of  $\text{SF}_6$  molecules in the strong field of a pulsed  $\text{CO}_2$  laser. An enrichment coefficient 2800 was obtained in one cycle without cascading.

1. We report in this article the first successful isotope separation by applying the molecular gas high-power IR pulses, with an exceptionally large enrichment coefficient in one cycle and with physical separation of the isotopes. The only known experiment on separation of  $^{10}\text{B}$  and  $^{11}\text{B}$  by applying high-power laser pulses to the  $\text{BCl}_3$  molecule<sup>[1]</sup> called for rapid chemical binding of the B atoms with  $\text{O}_2$  as an acceptor, and yielded an enrichment coefficient of about 10 during the stage of formation of the BO radical. The physical principle of separation in these experiments is based on selective chemically-irreversible dissociation of one of the isotopes of the  $\text{SF}_6$  molecule in the strong field of the IR resonant radiation of the laser. The described method seems to us the simplest method of all those published on laser isotope separation.

2. The experiment consisted of irradiating a cell with  $\text{SF}_6$  by focused radiation from a pulsed  $\text{CO}_2$  laser and recording the IR and mass spectrum of the residual gas ( $\text{SF}_6$ ) before and after the irradiation. The  $\text{CO}_2$  laser, with transverse discharge and preliminary photoionization, emitted pulses of frequency 1.5 Hz, energy 2 J, and duration at half-height 90 nsec. The generation frequency was tuned to the  $\nu_3$  oscillations ( $10.6 \mu$ ) with a dispersion resonator to the region of absorption of the individual isotopes of the  $\text{SF}_6$  molecule of natural isotopic composition. The laser beam was focused with a lens of  $f=50$  cm into a cell 50 cm long and  $60 \text{ cm}^3$  in volume, containing  $\text{SF}_6$  of natural isotopic composition ( $^{32}\text{S}-95\%$ ;  $^{33}\text{S}-0.75\%$ ;  $^{34}\text{S}-4.2\%$ ;  $^{36}\text{S}-0.017\%$ ), and in some cases acceptor molecules. The intensity of the IR field in the cell in the region of the focus, reached  $1-2 \text{ GW/cm}^2$ , i. e., the intensity at which collisionless dissociation of molecules is usually observed.<sup>[3]</sup> The isotopic composition was analyzed before and after the irradiation with the MI-1309 mass spectrometer. In addition, the remaining gas was condensed in a number of experiments after the irradiation in an additional cell, to investigate the degree of enrichment by measuring the IR absorption spectra in the  $10.6 \mu$  region.

3. When the frequency of the  $\text{CO}_2$  laser was tuned to the peak of the absorption band of  $^{32}\text{SF}_6$  at  $947 \text{ cm}^{-1}$ <sup>[3,4]</sup> (the P(16) line of the laser), the  $^{32}\text{SF}_6$  molecules disappeared almost completely from the mixture because of selective dissociation, after exposing the cell ( $\text{SF}_6-0.18$  Torr,  $\text{H}_2-2$  Torr) to  $2 \times 10^3$  pulses. The mass spectrum of  $\text{SF}_6$  contains mainly lines corresponding to the molecules  $^{32}\text{SF}_6$  and  $^{34}\text{SF}_6$ . From the ratio of the mass-line amplitudes before and after irradiation (Fig.

1) we obtain for the coefficient of enrichment of the mixture with  $^{34}\text{S}$  relative to  $^{32}\text{S}$

$$K\left(\frac{34}{32}\right) = \frac{[^{34}\text{S}]_k}{[^{32}\text{S}]_k} \bigg/ \frac{[^{34}\text{S}]_0}{[^{32}\text{S}]_0} = 2800$$

where the subscript "0" pertains to the non-irradiated mixture. Thus, the concentration of the  $^{34}\text{SF}_6$  molecules increased from 4.2 to 95%. The mixture is also noticeably enriched with the isotope  $^{33}\text{S}$  relative to  $^{32}\text{S}$ . Analogous experiments yielded a coefficient  $K(36/32) \approx 1200$  of mixture enrichment with the isotope  $^{36}\text{S}$  relative to  $^{32}\text{S}$ .

When the  $\text{CO}_2$  laser frequency was tuned to the absorption band of the  $^{34}\text{SF}_6$  molecule<sup>[3,4]</sup> (laser line P(40)) and the cell with the same composition was exposed to 30 pulses, an enrichment coefficient  $K(32/34) = 1.7$  was obtained for the  $^{32}\text{S}$  isotope relative to  $^{34}\text{S}$ , and  $K(32/34) = 18$  was obtained after 500 pulses. Thus the concentration of the  $^{32}\text{S}$  isotope in the mixture increased to more than 99%, while that of  $^{34}\text{S}$  decreased from 4.2 to 0.2%.

To illustrate further and demonstrate the physical separation of the isotopes, the residual gas was frozen out of the irradiated cell after the irradiation into a small cell suitable for the measurement of the IR spectrum. The initial cell was irradiated in such a way as to reconvert the  $^{32}\text{SF}_6$  molecules to the concentration of

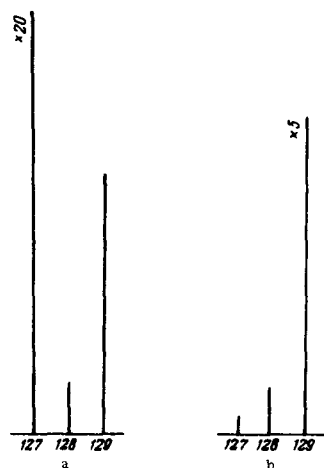


FIG. 1. Section of mass spectrum of  $\text{SF}_6$  (radical  $\text{SF}_6^+$ ): a) natural mixture prior to enrichment; b) after exposure to  $2 \times 10^3$  pulses of  $\text{CO}_2$  laser (P(16) line).

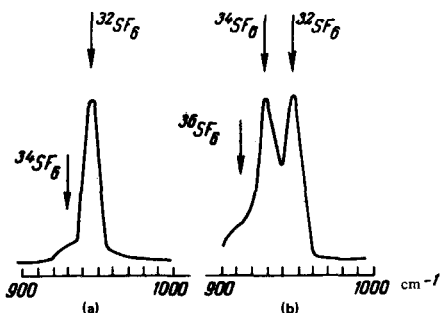


FIG. 2. IR absorption spectrum of the  $\nu_3$  oscillation in  $SF_6$ : a) natural mixture, b) mixture enriched in our experiments.

the  $^{34}SF_6$  molecules. Figure 2a shows the IR spectrum of the natural mixture of the  $SF_6$  molecules prior to irradiation in a cell of length 6 cm, volume  $25\text{ cm}^3$  and  $SF_6$  pressure 1 Torr. Figure 2b shows the IR spectrum after condensing in the same cell the residual  $SF_6$  mixture, after irradiating 0.5 Torr of the natural mixture of  $SF_6$  in a cell 100 cm long and  $500\text{ cm}^3$  in volume. It is clearly seen that the relative concentration of the  $^{34}SF_6$  and  $^{36}SF_6$  molecules has increased at the expense of burning out of the  $^{32}SF_6$  molecules.

We measured also the dependence of the enrichment coefficient  $K(34/32)$  at different  $SF_6$  pressures. The measurement results for various numbers of pulses are shown in Fig. 3. In the region below 0.5 Torr the enrichment decreases in inverse proportion to the pressure.

Qualitatively analogous results were obtained by irradiating both pure  $SF_6$  and following addition of  $H_2$ ,  $NO$ , and  $HBr$  as acceptors. A chemical analysis of the products of the dissociation and of the chemical reactions was not carried out, although the presence of  $HF$  was noted in the experiments with  $H_2$ .

5. As shown in<sup>[1]</sup>, a molecule located in a strong IR laser radiation field can enter selectively in chemical reactions. Although in our case, in contrast to<sup>[1]</sup>, the irreversible dissociation of the molecules took place

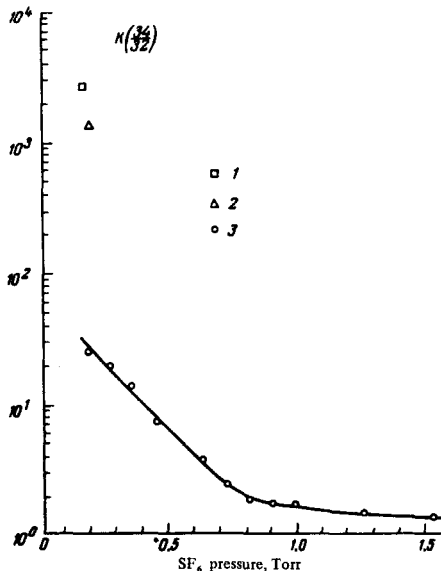


FIG. 3. Dependence of the enrichment coefficient of the isotope  $^{34}SF_6$  relative to  $^{32}SF_6$  on the initial  $SF_6$  pressure in the cell.

without visible luminescence, this is not a contradiction, but more readily a characteristic of radical formation in IR photolysis of the  $SF_6$  molecule. It seems that no radicals are produced in the electron-excited state.

Thus, we have first realized in our experiments a macroscopic separation of isotopes by IR laser emission, with an approximate yield of  $10^{-4}\text{ g}$  of  $SF_6$  and a 20-fold enrichment after 100 pulses of 2 J energy for one minute (at a repetition frequency 1.5 Hz).

<sup>1</sup>R. V. Ambartsumyan, V. S. Letokhov, E. A. Ryabov, and N. V. Chekalin, *ZhETF Pis. Red.* **20**, 597 (1974) [*JETP Lett.* **20**, 273 (1974)].

<sup>2</sup>R. V. Ambartsumian, V. V. Doljikov, V. S. Letokhov, E. A. Ryabov, and N. V. Chekalin, *Chem. Phys. Lett.* **25**, 1281 (1974).

<sup>3</sup>H. Brunet and M. Perez, *J. Mol. Spectr.* **29**, 472 (1969).

<sup>4</sup>V. D. Klimov and E. A. Lobikov, *Opt. Spektr.* **30**, 48 (1971).

**Erratum: Separation of sulfur isotopes with enrichment coefficient  $> 10^3$  through action of  $\text{CO}_2$  laser radiation on  $\text{SF}_6$  molecules**  
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On p. 171 the following callouts were omitted from Fig. 3: 1–200 counts (+ 2 Torr of  $\text{H}_2$ ), 2–400 counts, 3–100 counts.