Isotope Separation of $^{32}$SF$_{6}$ and $^{12}$CF$_3$I by a Q-Switched CO$_2$ Laser

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Abstract. Pulses with energy of 0.5 to 15 mJ and a repetition rate of 20 kHz from a Q-switched CO$_2$ laser with continuous discharge were employed to dissociate $^{32}$SF$_{6}$ and $^{12}$CF$_3$I at 0.25 mbar. Gas exchange by diffusion is rapid enough to maintain isotopic selectivity. Dissociation yields agree with those measured at slightly raised temperature by a TEA laser. Because photons are cheaper from Q-switched lasers than from TEA lasers, economic consequences can result for isotope separation.

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In a well-designed laser isotope-separation process, the costs depend primarily on the price of producing the photons, at least in the case of light elements and stable isotopes. The cost of laser light is dominated by the cost of the laser and by the number of photons it emits before it must be replaced. For isotope separation by infrared multiphoton dissociation lasers with pulsed discharges (e.g., TEA CO$_2$ lasers) have been used so far. Their lifetime is still unsatisfactory, whereas their average power begins to attain the range of interest for production plants [1]. Much longer lifetimes and therefore cheaper photons are customary for continuous wave CO$_2$ lasers, and kilowatt powers are commercially available. It would be economically interesting, if a large fraction of this power can be extracted in Q-switch operation and if this can be used for isotope separation.

We are developing such a laser. A preliminary version has pulses of 300 ns length and up to 20 mJ energy. The extraction efficiency requires a repetition rate of 10 to 20 kHz. Here we demonstrate that isotope selective dissociation of SF$_6$ and CF$_3$I is feasible by such lasers. To avoid a nonselective thermal reaction, the gas must be exchanged in the time (50 μs) between the laser pulses. This was not done by gas flow, but passively: Diffusion is fast enough at pressures below 0.3 mbar and under conditions of strong focussing (waist $w_0 \leq 0.1$ mm). But some thermal effects were observed, too: The steady-state temperature was around 390 K. The dissociation yields deviate from TEA laser data at room temperature, but agree perfectly with those at 390 K.

The deviation from the room-temperature data were unexpected, because pulse length and shape effects were known to be unimportant in these molecules at the energy densities under consideration [2, 3]. The lower yield seemed also a drawback for practical application. However, the agreement at the higher temperature supports the original expectation, that a Q-switched laser is an attractive alternative for a TEA laser in an isotope-separation plant; only a faster gas exchange would be necessary. It is true that in the present experiment the photons were not used efficiently, because of our single-focus conditions. How to use them economically was, however, described in [4].

Previously, multiphoton dissociation by a low-repetition-rate Q-switched CO$_2$ laser has already been observed [5] with perfluoro cyclobutane, a molecule which only requires low power; isotopic selectivity has not been investigated there.

Experimental Setup and Evaluation

Details about the laser can be found in [6]. Briefly it consists of an oscillator-amplifier arrangement. The oscillator is tunable by a grating and is electrooptically...
Q-switched by a Pockels cell. For the present experiments it was operated at a repetition rate of 20 kHz. Its pulses had energies up to 2 mJ, lengths of 300 ns half width and a smooth temporal shape. At the corresponding average power (30 W) the warming of the Pockels crystal (CdTe) was just tolerable, so that the beam quality still remained nearly diffraction limited.

In most experiments this radiation was amplified in a commercial laser (Ferranti MFK) with longitudinal discharge and slow longitudinal flow. It consists of a folded array of 12 tubes containing 24 m of gain length. If used as an oscillator, it can emit up to 1200 W of cw power. In the present experiment we used only half of these gain sections. We obtained pulses of 10 to 20 mJ (average powers up to 300 W) with 250 ns length. More than 80% of the power had diffraction limited quality. This was estimated according to the transmission through an iris with variable diameter.

The laser beam was focused into a cell of 100 mm length and 36 mm inner diameter (volume 102 cm³), equipped with NaCl windows, by means of a NaCl lens of 155 mm focal length. Before this lens the beam diameter and convergence was varied by a mirror telescope. Focal waists \( w_0 \) (e⁻² radius) in the cell varied from 0.03 to 0.1 mm and the peak energy density

\[
\phi_{\text{max}} = \frac{2E_p}{\pi w_0^2}
\]

(\( E_p \): energy of a laser pulse) ranged up to 300 J/cm² (calculated). For some experiments we used in the focus of the telescope an aluminum diaphragm which transmitted only the diffraction limited fraction (∼80%) of the beam.

Times of irradiation to 50% conversion ranged from several seconds to 30 min. The remaining fractions of SF₆ and CF₃I obeyed

\[
n_d/n_0 = \exp(-us),
\]

where \( n \) denotes the gas density; \( s \) the number of pulses ("shots"), and \( u \) the fraction of the cell volume converted per pulse. Equation (2) is valid for \( u \ll 1 \); in our case \( u \) was \( 10^{-6} \) to \( 10^{-4} \). \( n_d \) and \( n_0 \) were determined before and after irradiation by an infrared spectrometer: for \(^{32}\text{SF}_6\) and \(^{34}\text{SF}_6\) by the \( v_3 \) band and for \(^{12}\text{CF}_3\text{I} \) and \(^{13}\text{CF}_3\text{I} \) by the \( v_4 \) band. A quantity independent of the cell volume \( V \) is

\[
U = uv,
\]

which is the equivalent volume in which the converted molecules would be contained at a density \( n_0 \).

The conversion \( U \) (German: Umsatz) is a certain spatial average of the local dissociation probabilities \( P_d \). For a focused Gaussian beam the averaging has been performed in [7] for several different functional forms of \( P_d(\phi) \). The result factorizes into a geometrical part (which is kind of focal volume [7]) and a function \( Y(\phi) \), which only depends on \( P_d(\phi) \) and thus on molecular parameters

\[
U = Y(\phi_{\text{max}}) \cdot 2(\pi w_0^2)^{3/2}/\pi/3\lambda
\]

(4)

where \( \lambda \) is the wavelength.

So we measured for various pulse energies (with and without amplifier, different pressures in the lasers) and focussing conditions (thus various \( w_0 \) and \( \phi_{\text{max}} \)) the depletion \( n_d/n_0 \). After applying a temperature correction as discussed below, we deduced by (2–4) the experimental \( Y(\phi_{\text{max}}) \). This was compared with the \( Y \) based on dissociation probability functions \( P_d(\phi) \) of various shapes.

### Results and Discussion

Figure 1 shows the reduced experimental conversions \( Y \) of \(^{32}\text{SF}_6\) obtained from irradiations at the laser line 10P20. The curves have been deduced [7] from model functions of the dissociation probabilities \( P_d(\phi) \), one of them (broken line in Fig. 1) corresponding to the experimental \( P_d \) measured [8] by a TEA laser at the laser line 10P20. Figure 2 shows these \( P_d \) functions. Two alternative model \( P_d \) [Ref. 7, Eqs. (3 and 7)] are

![Fig. 1. Reduced conversion \( Y \), see (4). \( \phi_{\text{max}} \) is the maximum energy density in the focused Gaussian beam (1). Asterisks: with spatial filter. The broken line has been derived from TEA laser data at room temperature [8]. The curves are based on the Arrhenius-like functions [7] for the dissociation probability \( P_d \) with 50% point 15 J/cm² (both curves) and steepness parameter \( n = 1 \) (broken line) and \( n = 11 \) (solid line). For these functions see Fig. 2](image-url)