SHORT NOTE

Measurement of CO$_2$ Laser Pulse Narrowing in CF$_2$HCl

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It is important to study the characteristics of laser pulse propagation in order to scale-up the laser isotope separation (LIS) process, and to obtain efficient use of the available photon energy. During the laser propagation through an atomic medium, several nonlinear phenomena have been observed$^{(1)}$-$^{(3)}$. On a molecular medium, however, it is difficult to theoretically describe nonlinear effects because of the more complicated process of intramolecular relaxation and multiple photon absorption$^{(4)}$.

The experimental results and the theoretical models$^{(5)}$ were obtained for the transmission of laser pulse in gaseous SF$_6$ molecules in the case of weak excitation. In this experiment, the absorption energy was not sufficient to cause dissociation of SF$_6$. In the strong excitation, the narrowing of a CO$_2$ laser pulse due to the nonlinear photon absorption was observed$^{(6)}$ in CF$_2$HCl gas which is suitable for the carbon LIS process$^{(7)}$-$^{(9)}$. However its results was rather qualitative and was not sufficiently applicable to the analysis of laser propagation in the LIS process. In this study, quantitative measurements were performed to evaluate the dependence of pulse narrowing on the pressure of CF$_2$HCl and the laser wavelength.

1. Experiment

In Ref. (6), the experimental setup which was utilized has been described in detail. The CO$_2$ laser beam was not focused to avoid the effect of irradiation geometry. The laser-induced reaction of CF$_2$HCl is particularly simple, being represented by$^{(9)}$

$$\text{CF}_2\text{HCl} + n\hbar \nu \rightarrow \text{CF}_3 + \text{HCl}, \quad (1)$$
$$2\text{CF}_3 \rightarrow \text{C}_2\text{F}_4. \quad (2)$$

As HCl accumulates, reaction of CF$_3$ with HCl competes effectively with that by Eq. (2), and the net reaction rate decreases$^{(10)}$.

The laser pulse was measured at the cell outlet with a photon-drag detector connected to an oscilloscope (Sony Tektronix 2467B). The electric discharge of the laser pulse established a trigger signal for this measurement. The degree of photodissociation was measured with a gas chromograph.

In this experiment, a digitizing camera system (DCS) was utilized to record the waveform of output pulses on the CRT as a sequence of $X$-$Y$ data. This data set was analyzed by a computation program to obtain specific values describing a pulse reshaping. The pulse FWHM (full width at half maximum) and FWQM (full width at quartered maximum) were selected to represent a pulse reshaping. The latter represents the effect of photon absorption at both the leading edge and the tail part. A spatial filter of 6 mm diameter was utilized to avoid the effect of gas decomposition by the multiple photon dissociation because the cell pressure was varied with decomposition. In the measurement of a decomposition fraction, the spatial filter was removed.

2. Results and Discussion

The dependence of the pulse FWHM and FWQM on the pressure of CF$_2$HCl are shown in Fig. 1 (a) and (b) for 9R26 (pulse energy of 2.80 J), 9R20 (3.15 J) and 9R14 (3.45 J) lines, respectively. The decrease of the pulse FWHM and FWQM with the increase of pressure was observed in all figures. The decrease of the FWQM is observed rather significant than the FWHM in all cases. This means that the absorption at leading edge and pulse tail is stronger than the absorption at a pulse peak. Therefore the photon absorbing ability of a molecule is dependent on the (instantaneous)

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laser intensity. As the laser is tuned near resonant, the decrease of pulse width became significant. For 9R26 line at 60 Torr, the FWHM and FWQM decreased to almost the half of an input pulse.

The decomposition fraction due to the multiple photon dissociation was measured after irradiation of 600 pulses. Figure 2 shows the dependence of the decomposition fraction on the pressure for three different oscillation lines, 9R26, 9R20 and 9R14. As the laser is tuned off resonance, the decomposition fraction decreases. Although the wavenumber is different, the difference of decomposition between 9R26 and 9R20 is very little. The reason of this results can be considered as follows; (1) a pulse energy of 9R20 is larger than 9R26 resulting in the higher fluence, and (2) the accumulation of HCl enhance the recombination of photodissociated fragments and there may exist the upper limit of the decomposition fraction. The reason (2) can be also concluded from the fact that the decrease of the FWHM and FWQM is rather significant for 9R26 line although the decomposition fraction is almost as the same as 9R20 line.

As the pressure is raised, the decomposition is enhanced in low pressure range. However the decomposition fraction decreases in accordance with the increase of the pressure in relatively high pressure range. The increase of the decomposition fraction in low pressure range can be explained by the collision broadening of each vibrational level. The collisional broadening of levels will compensate for the anharmonic defect of a level separation. It is rather difficult to explain the decrease of the decomposition fraction in high pressure range. One of the reasons may be that the collisional de-excitation overcomes the broadening of vibrational levels. Furthermore there is a threshold intensity for photodissociation and the dissociation yield is nonlinearly dependent on the fluence. If a pulse intensity is reduced below the threshold due to the nonlinear absorption, a pulse has no ability to dissociate molecules. Additionally, the dissociation yield
suddenly decreases with the decrease of the fluence. Therefore the too high pressure may decrease the effective reaction volume. Accordingly the pressure corresponding to the maximum decomposition fraction may also depend on the cell length.

The $^{13}$C concentration of the product $\text{C}_2\text{F}_4$ was also measured in all cases. In this experiment, however, a significant enrichment was not observed. For the enrichment of $^{13}$C, the optimum oscillation line is $9\text{P}20^{(7)}\sim^{(9)}$ which has a much longer wavelength than those utilized in this experiment. Therefore the absorption cross section of $^{13}$C can be assumed almost as the same as $^{12}$C on the condition of this experiment.

3. Conclusion

The nonlinear absorption of a $\text{CO}_2$ laser pulse resulting in a decrease of the pulse FWHM was quantitatively observed in this experiment. The nonlinear absorption took place during conditions in which multiple photon dissociation was induced. This pulse reshaping became significant when the pressure of CF$_2$HCl was raised or the $\text{CO}_2$ laser was tuned nearly resonant. In evaluating the effective optical length, this nonlinear photon absorption should be taken into account.

References