SHORT NOTE

Experimental Study of Absorption Characteristics of CO₂ Laser Pulse in CF₂HCl

Kazuyuki KATOH, Hiroshi SUGAI and Atsuyuki SUZUKI

Department of Nuclear Engineering, University of Tokyo*

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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. Several studies have previously reported the existence of non-linear phenomena such as self-focusing, self-defocusing, self-steepening and frequency modulation that can be observed during the laser propagation process(1)~(3). Using an atomic rubidium medium these nonlinear phenomena effects were investigated not only experimentally but also theoretically(4). On a molecular medium, however, it is difficult to theoretically describe these effects because of the more complicated processes of intramolecular relaxation and multiple photon absorption(4).

The experimental results and the theoretical models proposed in Ref. (5) for the transmission of laser pulses in gaseous SF₆ molecules were obtained by weak excitation, thus only a few photons were absorbed and the absorption energy was not sufficient to cause dissociation of SF₆. As a result, these results cannot be applied to the LIS process which utilizes multiple photon dissociation of molecules.

In this experiment, CF₂HCl gas was utilized as an absorbing medium because it is suitable for the carbon LIS process. Due to the low critical fluence of CF₂HCl, multiple photon dissociation is easily induced to cause strong photon absorption. Reference (1) indicated that significant pulse reshaping did not occur if the laser pulse was strongly absorbed. An experimental study was performed to observe the time dependent behavior of a CO₂ laser pulse where strong excitation lead to multiple photon dissociation.

1. Experimental Setup

Figure 1 shows a schematic diagram of the experimental setup which was utilized. The line-tunable CO₂ laser (Lumonics TE-821HP) produced a 3 cm × 3 cm square laser beam with a pulse energy of several joules. A cell containing CF₂HCl was 45 cm long and had Ge windows coated with Au-Cu on both sides to minimize the reflection losses. The CF₂HCl pressure was measured by a quartz vacuum gauge (ULVAC GX-1) and was varied from 0.09 to 300 Torr. The cell was heated to approximately 373 K to avoid condensation of the HCl that is produced by the dissociation of CF₂HCl. Laser wavenumbers were determined with a spectrum analyzer (Optical Engineering 16-A). The laser pulse was measured at the cell outlet with a photon-drag detector (HAMAMATSU B749; maximum rise time of 1ns) connected to a storage type oscilloscope (Hewlett-Packard 1744A). The output pulse was photographed directly on the CRT of the oscilloscope. The electric discharge of the laser pulse established a trigger signal for this measurement. Photon dissociation was measured with a gaschromatograph (SHIMADZU GC-8A).

2. Results and Discussion

The infrared absorption spectrum of CF₂HCl(6) is shown in Fig. 2 for a weakly excited state. The region indicated between the dotted lines corresponds to the available wavenumber range of the CO₂ laser oscillation. The line numbers 9R32, 9R10 and 9P8 were examined in this experiment as the cell pressure and the input pulse energy were varied. The CO₂ laser was operated at a discharge voltage of 34 kV, so that the pulse energy was dependent on an oscillation line. A mesh

* Hongo, Bunkyo-ku, Tokyo 113.
The sequence of output pulses shown in Fig. 3 shows the pressure dependent absorption characteristic of 9R32 line for a pulse energy of 0.4 and 1.5 J. As the pressure of the cell was raised from 0.09 to 20 Torr, it was determined that the maximum output pulse peak height to maximum output pulse tail height ratio nonlinearly increased due to photon absorption. As a result of this nonlinear absorption the full width at half maximum (FWHM) of the pulse duration was also reduced. This phenomenon is similar to saturable absorption. As the laser input energy was increased from 0.4 to 1.5 J the max peak to max tail ratio also increased, although not significantly. After the irradiation of 1,000 pulses using the above conditions the gas-chromatograph was used to examine whether multiple photon dissociation was induced or not. It was observed that approximately 50% of molecules contained in the cell were dissociated in case of the strong excitation. In case of the weak excitation (0.4 J), however, over 20% of molecules were also dissociated because the 9R32 line was on resonance.

The pressure dependent absorption characteristics of line numbers 9R10 (pulse energy of 2.7 J) and 9P8 (2.3 J) are shown in Fig. 4. The 9R10 line is also considered to be near resonant as a result of the red shift in the absorption spectrum which occurs at strong excitation. Therefore, similar to the 9R32 absorption, nonlinear absorption which results in a decrease of pulse FWHM and an increase of the max peak to max tail ratio was observed for 9R10. For the 9R10 it was found that about 7% of molecules were dissociated. On the other hand, nonlinear absorption was not observed for the 9P8 line which was far off resonant and the amount of dissociation was negligible.

It is postulated from the experimental results that nonlinear absorption takes place under conditions where multiple photon dissociation is induced. As a laser pulse propagates through a molecular medium, its energy and the FWHM of the pulse decrease. Nagi et al. measured the laser power dependence of the absorbed photon number per molecule of C₂F₅Cl. Their result indicated that the absorbed photon number was directly proportional to laser input power below the
dissociation threshold, and that above the dissociation threshold the absorbed photon number was proportional to the square root of the laser input power. Thus the photon absorbing ability of a highly excited molecule is different from a weakly excited one. This same difference in molecule absorption ability played an important role for this experiment with respect to the absorption characteristics of a CO$_2$ laser pulse propagating through a molecular medium of CF$_2$HCl. The nonlinear absorption which was observed may be caused by the saturation of quasi-continuum levels of the molecule.

3. Conclusion
The nonlinear absorption of a CO$_2$ laser pulse resulting in a decrease of the pulse FWHM was observed in this experiment as evidenced by max peak to max tail ratio. 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 CO$_2$ laser was tuned nearly resonant. Additionally the peak intensity of the pulse was also reduced by the absorption.

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