Identification of a narrow resonance structure of energy absorption by sulfur hexafluoride molecules in a strong IR laser field

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A high-pressure, continuously tunable CO_2 laser is used to identify narrow peaks of the resonant absorption by SF_6 molecules in a strong IR laser radiation field. These peaks are interpreted as two-photon absorption peaks in the system of lower vibrational levels.

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In the collisionless dissociation of multiatomic symmetric molecules the intramode anharmocity of several lower vibrational levels can be overcome, according to the existing theoretical ideas,[1,2] in the process of two- and three-quantum transitions in the level structure that is formed as a result of de-excitation of the excited levels due to anharmonicity. This structure has narrow resonance properties whose presence is essential for spectral selectivity of the dissociation process. An attempt to observe experimentally such a structure was made[3,4] by using atmospheric-pressure CO_2 lasers whose emission frequency was discretely tunable for the emission lines with an interval ~ 1.5 cm^{-1}. The results[3,4] showed that the resonance structure of the absorption of energy by symmetric multiatomic molecules in a strong IR laser field with continuous frequency tuning must be investigated.

In this paper we report the observation of sharp peaks of the resonance absorption of laser energy by SF_6 molecules with the help of a high-pressure CO_2 laser. We used a laser with a non-self-sustained discharge which was electron-beam-assisted at a pressure of 6 atm. The design of this laser was described in Ref. 5. The energy reached 1 J/cm^2, the pulse duration was 40 nsec, the spectral width of emission was 0.03 cm^{-1}, the total range of continuous tuning was 86 cm^{-1}, and the tuning increment was 0.06 cm^{-1}.

The energy absorbed in the gas was measured optically and acoustically. To eliminate the effect of hot bands of the cell the gas mixture (SF_6:Xe = 1:10) was cooled to 140 K. The total pressure was 0.3 Torr. To lock the frequency, we measured the absorbed energy in the CO_2 gas in which the location of the absorption lines is well known. The presssure in the cell containing CO_2 was 1.5 atm. Figure 1 shows the absorption spectra of SF_6 molecules at densities of irradiated energy of 0.09 J/cm^2 (lower curve) and 0.27 J/cm^2 (upper curve). The upper part of Fig. 2 shows the absorption spectrum of CO_2 in which the contrasting absorption peaks are observed. The width of the peak at the frequency 944.46 cm^{-1} is 0.25 cm^{-1}. Figure 2 shows the dependence of the absorbed energy E_{abs} on the density of the irradiated energy Φ for the two most intensive peaks at the frequencies 944.46 cm^{-1} and 947.98 cm^{-1}. In

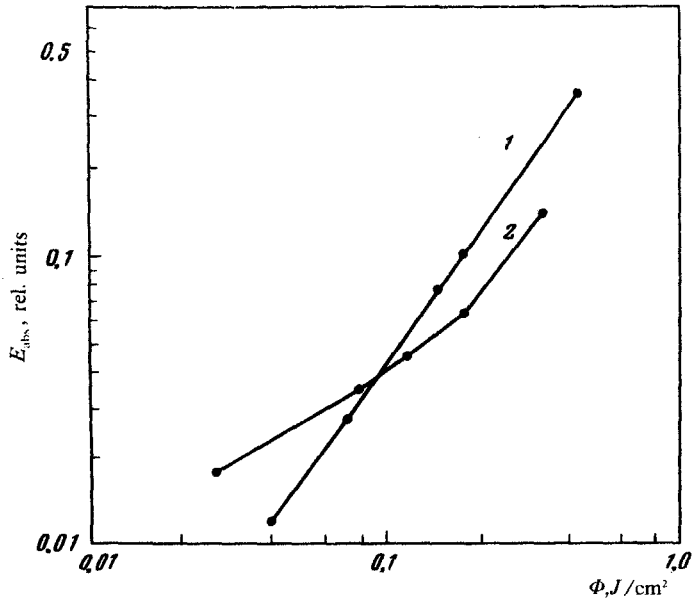

FIG. 1. Dependence of the energy absorbed in SF_6 (E_{abs}) on the energy density of the laser field Φ at the excitation frequencies 944.46 cm^{-1} (curve 1) and 947.98 cm^{-1} (curve 2).

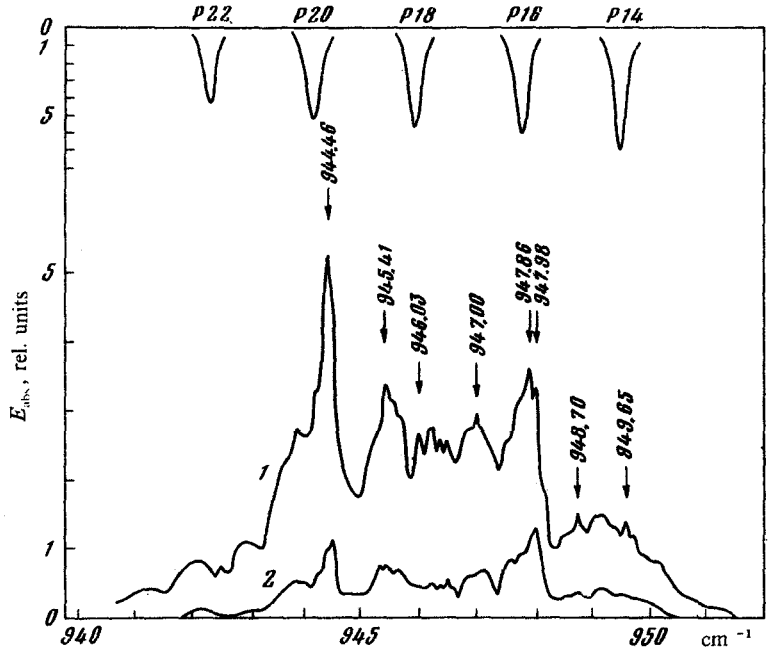

FIG. 2. Dependence of the energy absorbed in SF_6 on the frequency of the laser field at excitation-energy density 0.27 J/cm^2 (curve 1) and 0.09 J/cm^2 (curve 2). The pressure is 0.3 Torr, the temperature is 140 K, and the resolution is 0.06 cm^{-1}. The upper part shows the energy absorbed in CO_2 at the pressure of 1.5 atm.

and 947.98 cm^{-1}. In the region of small energy densities at the frequency 947.98 cm^{-1}, the dependence of the absorbed energy E on the density of the irradiation energy Φ has the form $E \sim \Phi^{0.5}$ which indicates that the single-photon absorption near the Q branch of the main 0–1 transition is the dominant contribution. For the peak at the frequency 944.46 cm^{-1} this dependence has the form $E \sim \Phi^{1.65}$ in the entire range of investigation. This peak, is apparently connected with the Q branch of the two-photon transition to one of the sublevels of the vibrational state $v = 2$. The frequency position of this peak and the data on the frequency of the $3\nu_3 SF_6$ harmonic[6,7] lead us to conclude that the anharmonic splitting in SF_6 is large.

The observed structure, which apparently is common in molecules of this class, can be used to improve the selectivity of excitation of heavier molecules.

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