

NONLINEAR SPECTROSCOPY: EXPERIMENTS WITH MOLECULES IN A TWO COLOUR RESONANT IR RADIATION FIELD AND THE THEORETICAL MODELLING BASED ON THE OPTICAL BLOCH EQUATION.

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Recent achievements in nonlinear optics opened new possibilities in IR high resolution spectroscopy. In this contribution a two colour spectroscopy of cooled SF₆ and C₂H₄ seeded in a He beam is presented. The molecules were excited by CW CO₂ laser beams crossing the molecular beam. The frequencies of laser beams were tuned around CO₂ lines and shifted by Optoacoustic Modulators (OAM) in the range ± 500 MHz. The spectral resolution of 2 MHz was achieved. The excitation spectra were modelled in the dressed state picture (DSP) and by optical Bloch equations (OBE).

In C₂H₄ the one quantum transition ($v = 0, \{4, 1, 3\} \rightarrow v = 1, \{5, 0, 5\}$) was investigated. At the moderate intensities of the laser fields (ν_1 and ν_2) the UP-DOWN-UP-... (UDU) scheme of excitation was realized. The peaks with the frequencies ν_1 and ν_2 satisfying the resonant condition $(N + 1) \cdot \nu_1 - N \cdot \nu_2 = \nu_{transition}$ with N=1 and N=2 were observed. The AC-Stark shifts of peaks were measured being in agreement with the results of computer modelling based on OBE. The modelling showed that increasing the laser intensities leads to a transition from the UDU picture to the AC-Stark split one photon line picture.

In SF₆ the two photon ladder-type transition ($v_3 = 0, J = 4 \rightarrow v_3 = 1, J = 3 \rightarrow v_3 = 2, J = 3$) was investigated. The frequency ν_1 was varied near the frequency of the first transition while ν_2 was varied near to the frequency of the second transition and to the frequency of the two photon transition.

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