TWO-COLOR RESONANT FOUR-WAVE MIXING SPECTROSCOPY OF HIGHLY PREDISSOCIATED LEVELS IN THE A^2A_1 STATE OF CH_3S

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We report results of two-color resonant four-wave mixing experiments on highly predissociated levels of the methylthio radical, CH_3S in its first excited electronic state. The radical was generated by 248 nm photolysis of dimethyl disulfide, and the spectra measured in a hole-burning scheme in which the probe laser excited specific rotational transitions in band 3^3. The spectral simplification afforded by the two-color method allowed accurate determination of line positions and homogeneous linewidths, which are reported for the C-S stretching states 3^n with n=3-7 and combination states $1^1 3^n$ (n=0-2), $2^1 3^n$ (n=3-6), and $1^1 2^1 3^n$ (n=0,1). The spectra show pronounced mode specificity, as the homogeneous linewidth of levels with similar energies varies by up to two orders of magnitude. The derived vibrational parameters of the A^2A_1 state are in satisfactory agreement with *ab initio* predictions.