

ANTICROSSING IN PROPYNAL EXCITED BY A NEAR U.V. HIGH REPETITION RATE COPPER VAPOR LASER PUMPED DYE LASER

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ANTICROSSING IN PROPYNAL EXCITED BY A NEAR U.V. HIGH REPETITION RATE COPPER VAPOR LASER PUMPED DYE LASER

J.P. PIQUE, M. LOMBARDI, U. BRÜHLMANN* and H.R. HUBER*

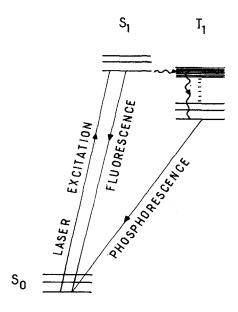
Laboratoire de Spectrométrie Physique de l'USTMG, CNRS UA-08 et Greco Celphyra, BP 87, F-38402 Saint-Martin-d'Hères Cedex, et SNCI LP 2031 et Greco Celphyra/CNRS, BP 166 X, F-38402 Grenoble Cedex, France
*Phys. Chem. Inst. Universität, Wintherturerstrasse 190, CH-8057 Zürich

1) PRINCIPLE of THE ANTICROSSING EXPERIMENT.

The molecule to be studied (here

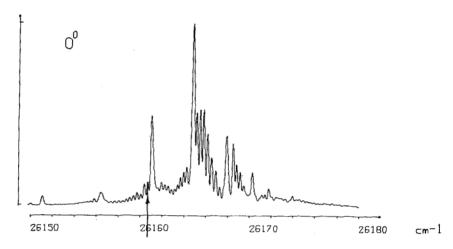
Propynal -> H-C\u00e4C-COH)

is seeded in an Helium supersonic beam which cools it to a few Kelvins without condensation.

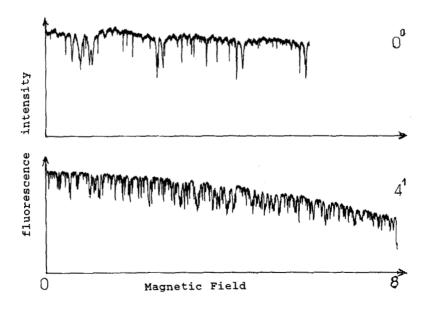


The molecule is excited to the first electronically excited singlet S_1 state by a Dye Laser

The fluorescence excitation spectrum is first recorded, giving a low temperature vibrationally and rotationally resolved spectrum.



Then the wavelength of the laser is held fixed on a given absorption line and Magnetic Field is swept (0 - 80000 Gauss). The singlet excited level remains unaffected by the field but neighbouring Triplets are displaced. Each time a triplet crosses the singlet, there is a dip in fluorescence (and a corresponding bump in phosphorescence not recorded in the present experiment).



The position of the dip gives the energy separation between singlet and triplet rotational levels, enabling full spectroscopy of the triplet which is impossible by conventional optical spectroscopy due to extreme weakness and congestion of the direct S_0-T_1 intercombination bands with such vibrational energy within the triplet. The width of the dip gives the coupling strength.

2) EXPERIMENTAL PROBLEMS.

Since signals are dips on the fluorescence intensity background, this experiment is rather sensitive to amplitude fluctuations of the laser. The problem is much less severe when recording spectra, where 20% amplitude stability is usually well enough, or when measuring lifetimes with a fast transient recorder which digitizes the whole decay curve on each laser pulse, but it is the key problem in the present experiment. First experiments were done with a cw laser, but this limits nearly to the visible spectrum, whereas most molecules have electronic absorption in the U.V. Common U.V. pulsed lasers (excimer or YAG pumped), are hampered by low rep rates and high peak to peak fluctuation intensities of the lasers. The high repetition rate of the Copper Vapor Laser (6 kHz) makes easy to average amplitude fluctuation to reach the ultimate sensitivity limit given by the photon noise. Since commercial dye lasers for Copper Vapor lasers were not available we have built one (of the Litman principle).

3) PHYSICAL INTEREST

In previous cw experiments using glyoxal (COH-COH), it has been found that the strength of the S_1 - T_1 coupling does not decrease when increasing energy, as was predicted by usual theories. We have made this experiment to check whether this is some peculiarity of glyoxal or a general rule. Present preliminary experimental results seem to confirm the glyoxal findings.