Picture Processing

Two-Dimensional Digital Signal Processing I

Linear Filters

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Doppler-Free Two-Photon Spectroscopy of Polyatomic Molecules

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Two-photon spectroscopy has been shown to be a new spectroscopic method which yields spectral information complementary to the results of conventional spectroscopic methods such as one-photon absorption. This has been demonstrated some years ago by the gas phase two-photon spectrum of benzene [1]. Another virtue of the two-photon absorption process is that it can eliminate the Doppler-broadening under suitable experimental conditions. This has been predicted in 1970 by Vasilenko et al. [2] but the experimental realization has so far been restricted to the case of atoms and diatomic molecules whose spectra are relatively simple and consist of a single or only a few lines at most within the Doppler-width.

For larger molecules like benzene, where many rovibronic $S_1 \leftarrow S_0$ transitions are hidden within the Doppler-width of 1.7 GHz, the Doppler-free spectrum is supposed to yield numerous new informations about molecular structure and dynamics.

The comparatively low transition moment of benzene and the fact, that in thermal equilibrium more than 10^4 states are populated make an experiment with a single-mode *cw* dye laser (as commonly) used in Doppler-free high-resolution spectroscopy of atoms) barely possible.

Therefore, in a first experiment [3], only one of the two laser beams used was from a dye laser and the other one from a high power single-mode Ar^+ laser. Due to the slightly different frequencies of the lasers there remained a residual Doppler-broadening of about 50 MHz. The resolution is, however, sufficient to resolve the individual rotational transitions. Part of the 14_0^1 vibronic band recorded in this way is shown in Fig. 1.

With the experimental set up shown in Fig. 2 the efficiency of the two-photon transition can be considerably improved and not only

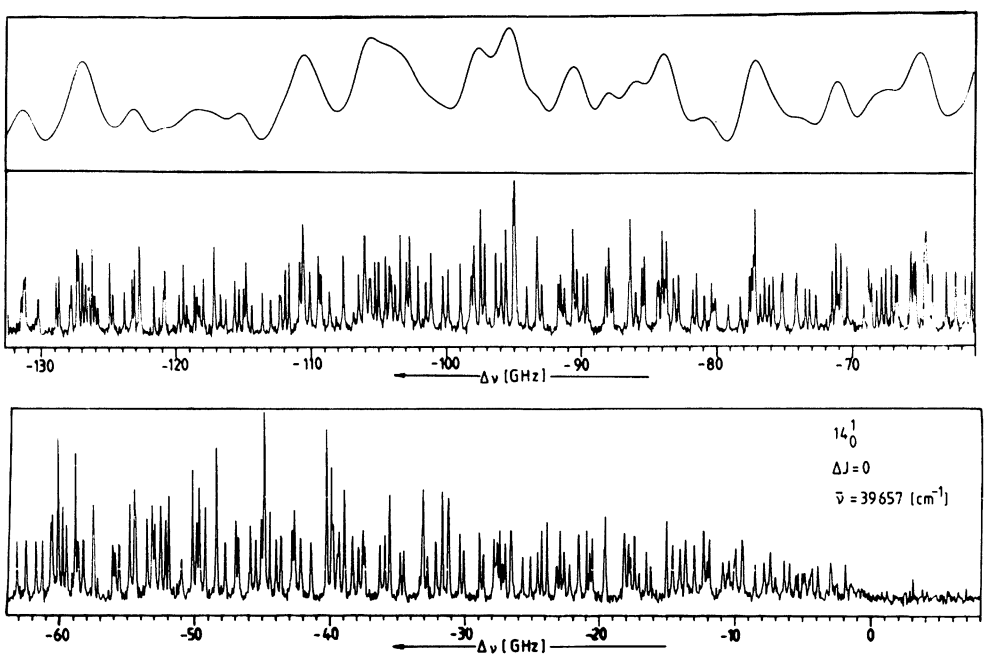

Fig. 1. Part of the Doppler-free two-photon spectrum of the benzene molecule. For comparison in the upper trace the spectrum of the middle trace is folded by the Doppler-width

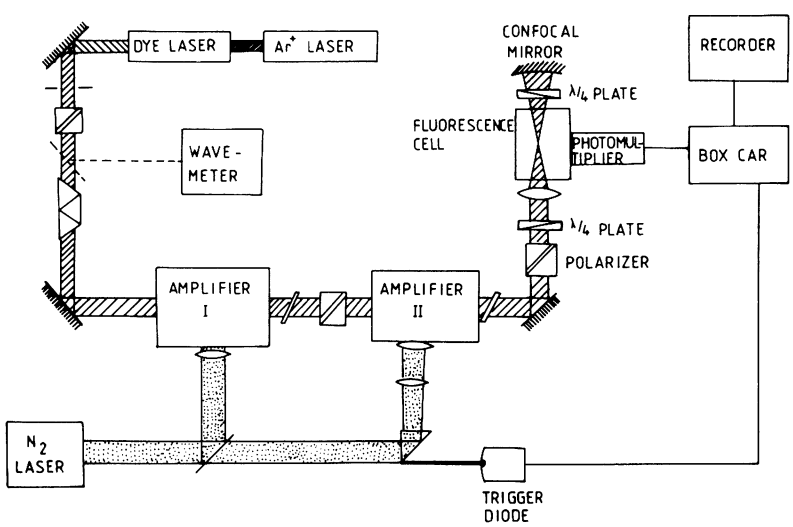

Fig. 2. Experimental set-up for Doppler-free two-photon spectroscopy with pulsed amplified light from a single-mode cw laser

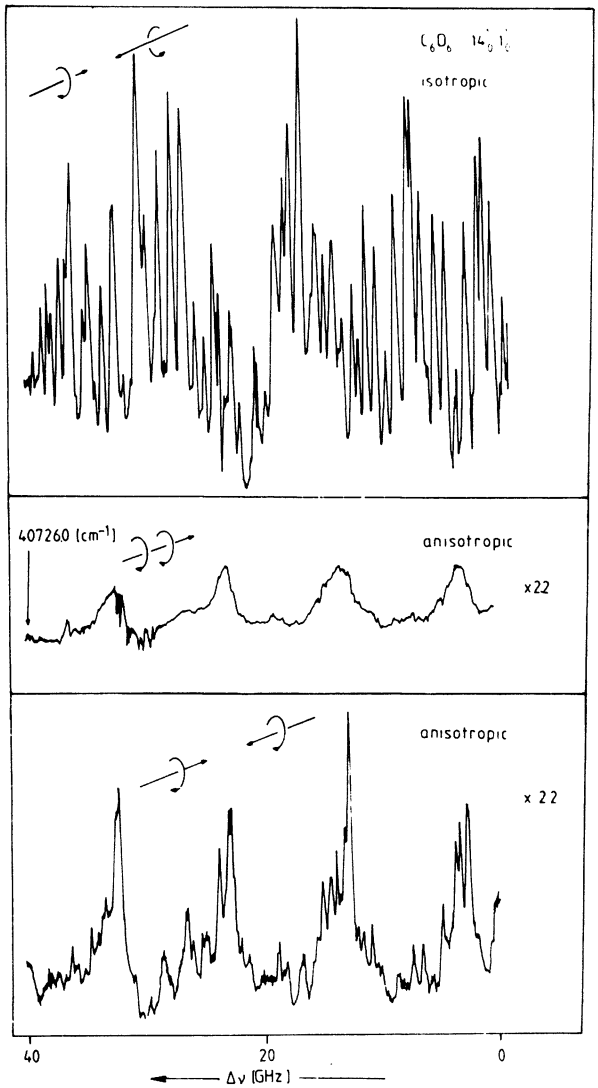

Fig. 3. Doppler-free (upper and lower trace) and Doppler-limited (middle trace) two-photon spectrum of benzene measured with the amplified pulsed light under different polarization conditions

the most intensive parts of the strongest vibrational bands can be investigated.

The single-mode *cw* dye laser was pulsed amplified in two stages pumped by a N_2-laser. Thus tunable radiation of about 50 kW and a nearly Fourier transform limited band width was obtained. This light was then focused into the fluorescence cell containing benzene vapor.

A typical part of the spectrum obtained in this experiment is shown in Fig. 3. In the middle trace only circularly polarized light from one beam was used giving a Doppler-broadened anisotropic spectrum. For recording the lower trace the laser beam was reflected back into itself by the confocal mirror. Now in addition Doppler-free anisotropic lines are observed based on a strong Doppler-broadened background due to the many lines within the Doppler width. By inserting a $\lambda/4$ plate between the cell and the mirror the sense of the circular polarization of the reflected beam is changed and the strong isotropic contributions of the spectrum can be seen [4] (upper trace) in addition to the unchanged and much weaker Doppler-broadened background produced only by the anisotropic part of the two-photon transition. Hence, this polarization behaviour is shown to be a facile method for suppressing the strong Doppler-broadened background of a polyatomic molecule.

As first results of our experiments we found a hitherto unexpected relatively large inertial defect of the benzene molecule. The explanation of this defect is obscured by the low resolution of ground state measurements, which can not decide the question of an inertial defect in the ground state of benzene.

We were furthermore able to measure collisional linewidths down to a pressure of 1 Torr. This gives for the first time the collisional parameter of a electronically excited polyatomic molecule. In the zero pressure regime the homogeneous linewidth is expected to provide us with information about the intramolecular dynamics. Additionally the pulsed setup gives us the possibility of measuring population lieftimes of single rotational states even in large molecules.

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