HIGH RESOLUTION LASERSPECTROSCOPY AS A DIAGNOSTIC TOOL IN BEAMS

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The combination of high resolution laser spectroscopy with the technique of molecular beams allows a very detailed beam research since molecules or atoms in specific quantum states can be sampled, yielding previously unavailable sources of data.

In our experiments a Na/Na_2 beam emerges from a 0.2 mm nozzle and is collimated by a 2 mm wide slit 50 cm downstream. To probe the molecules we used a single mode Ar^+-laser which can be tuned within the gain profile of the laser line (8 GHz) to several transitions between specific levels in the ground state and second electronically excited state of the Na_2 molecule (see Fig. 1). The quantum numbers of the participating levels are known from spectroscopic analysis[1]. The atoms are probed with a dye laser.

Monitoring the velocity distribution in a beam via Doppler-shifted emission was one of the first spectroscopic methods used in beams[2]. In the case of heavier molecules (e.g. Na_2), however, not only a high resolution Fabry-Perot interferometer but also a monochromator of medium or even high resolution is needed to select one single line of the emission spectrum. Another version of this technique makes use of the Doppler-shifted absorption via laser induced fluorescence[3,4]. The laser crosses the molecular beam perpendicular or nearly parallel for probing the perpendicular or parallel velocity distribution respectively. A photomultiplier detects the total fluorescence. If the laser is tuned across the absorption profile only molecules with a specified velocity can absorb. The fluorescence intensity $I(\Delta\nu)$ yields the velocity distribution since it is proportional to the number density $n(v)$. This is true only for sufficiently low laser intensities below the level where power broadening of the absorption line indicates depopulation of the ground state. The velocity v and the frequency offset $\Delta\nu$ are correlated via the Doppler-effect $v = \frac{c}{\cos\beta}\frac{\Delta\nu}{\nu_0}$ (ν_0 is the center frequency of the transition, β is the angle between the laser and molecular beam). A disadvantage of this method is that only the component of the velocity along the laser beam axis can be determined. This results in a correlation of the measured parallel and perpendicular distribution. Since the width of the latter distribution is dif-

ferent for molecules in different quantum states (see subsequent paper) high resolution in parallel velocity can only be achieved with highly collimated beams.

No such problems are encountered using the TOF via optical pumping method[5,6]. Even with moderate laser intensity considerable depletion of the population in a specific level occures through optical pumping in a beam due to the lack of collisional repopulation. The depopulation is demonstrated in Fig. 2. The pump laser crosses the beam vertically. A probe laser was used to monitor molecules in the pumped level passing through a 0.1 mm slit downstream of the pump laser. This analyzer was rotatable in the horizontal plane since we used fiber bundels for the probe laser as well as for collecting the induced fluorescence. A schematic set up of the TOF experiment is shown in Fig. 3. The pump laser is switched off with a rapidly rotating disk intersecting the laser beam in the focal plane between two lenses for about 30 μs. During this time, a group of molecules or atoms in the level under consideration can pass the pump laser without being optically pumped. These particles will arrive at the probe laser after a time delay according to their velocity, thus the time dependence of the fluorescent signal yields the TOF spectrum of molecules or atoms in a specific quantum state (see Fig. 4). In our case the probe laser also has sufficient power to cause considerable pumping. Each molecule passing through the analyzing beam has a chance close to unity for absorbing a laser photon. Then, in contrast to the Doppler-shifted absorption method, the detection efficiency is independent of the velocity and we are monitoring the flux of particles in the beam.

References

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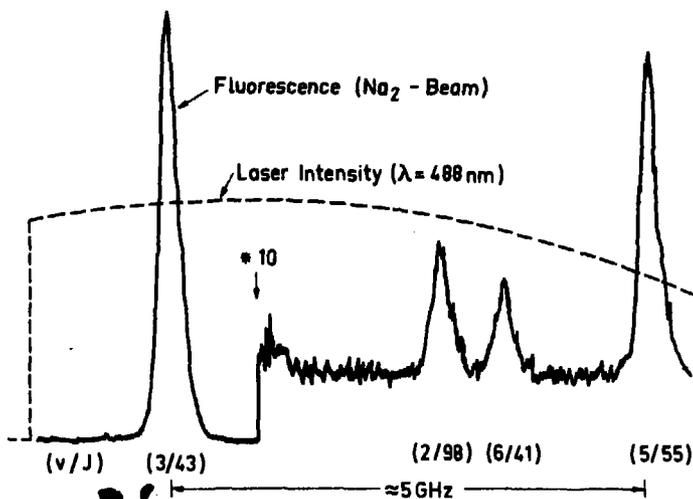6) K. Bergmann, U. Hefter, P. Hering, J. Chem. Phys. 65, 488 (1976)

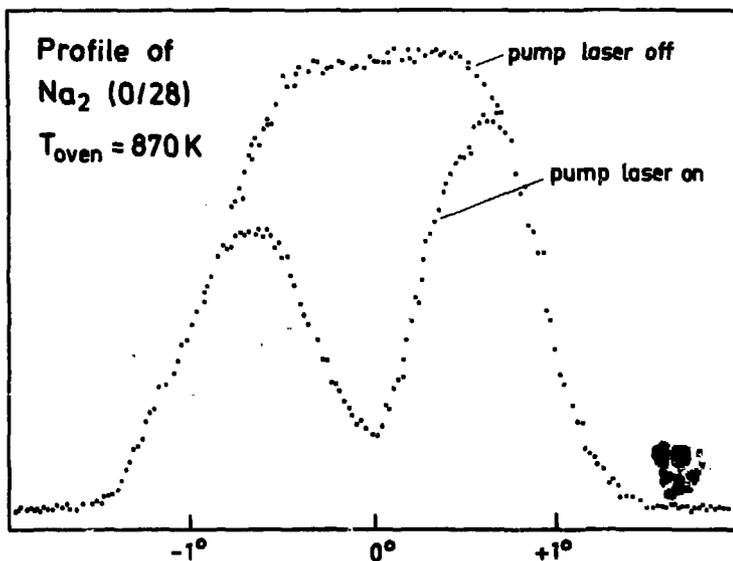Fig. 1: Spectrum of the total Na_2 fluorescence in part of the tuning range of the 488 nm Ar^+ laser line.

Fig.2: Beam profile of molecules in the (0/28) level with and without optical pumping. A probe laser detects the molecules behind a 0.1 mm slit. Fiber bundels are used for the probe laser as well as for collecting the fluorescence. The analyzer is rotatable in the horizontal plane.
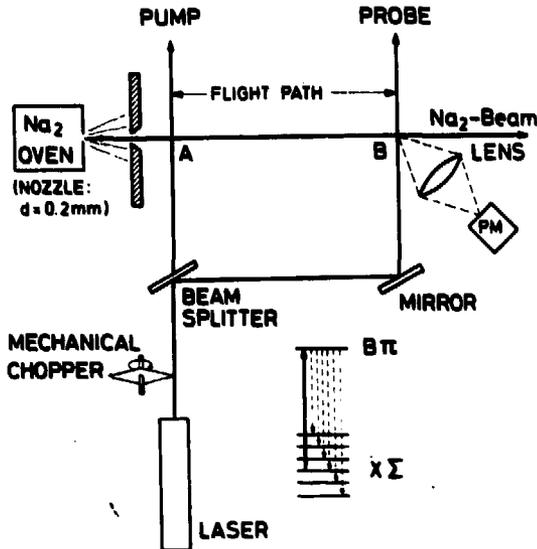
Time of Flight via Optical Pumping

Fig. 3: Schematic view of the set up for state selected time of flight experiment.

Fig. 4 (below): TOF spectrum for atoms in the lower hyperfine level of the $^2S_{1/2}$ ground state. A fit to the experimental data is also shown using

$$F(t) \sim \frac{1}{t^5} e^{-\beta^2 (\frac{L}{t} - u_a)^2}$$

which corresponds to

$$F(v) \sim v^3 e^{-\beta^2 (v - u_a)^2} \quad .$$

exp.
fit

Na-atoms
$T_{oven} = 10^{?} K$