Depolarization of NaD_2 Fluorescence under Pulsed Laser Excitation

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The intensity and polarization state of the NaD_2-fluorescence ($3\,^2P_{3/2} \rightarrow 3\,^2S_{1/2}$, λ 5890 Å) from a cell containing pure Na vapor, selectively excited by a flash lamp pumped dye laser was investigated. Measurements were performed both for excitation with π- and σ-light of spectral density ranging between 10^{-23} and 10^{-17} Ws^2 cm^{-3} and with Na number densities between 2.8×10^9 and 4.0×10^{10} cm^{-3}. The results are interpreted in terms of population of the $^2P_{3/2}$ state and of hyperfine and Zeeman pumping of the $^2S_{1/2}$ substates. The values for the degree of linear polarization measured at the smallest excitation densities and Na pressures agree well with theoretical values.

1. Introduction

The study of resonance fluorescence of gases or gas mixtures yields many informations on atomic structure as well as on interactions between atoms [1] or with the radiation field. In particular, if the exciting radiation is polarized, processes like collisional depolarization or transfer of polarization may be investigated [2]. With a dye laser as exciting light source it has now become possible to study collisions with atoms in highly excited or metastable states by stepwise or two photon excitation [3]. Also, using pulsed excitation, the time dependence of collisional energy transfer may be examined [4]. Finally, in principle not only total but also differential cross-sections of all kinds of collision processes may be investigated. However, the use of laser sources for these purposes requires at first consideration of effects of high radiation fields on the intensity and polarization state of the fluorescence radiation. This will be the topic of the paper presented.

In the following we report on fluorescence experiments using a pulsed dye laser for excitation. The Na transition $3\,^2P_{3/2} \rightarrow 3\,^2S_{1/2}$ (NaD_2, λ 5890 Å) was investigated at pure Na vapour in a resonance cell with number densities between 2.8×10^9 and 4.0×10^{10} cm^{-3}. The fluorescence was excited either with π or σ-polarized light of spectral densities variing between 10^{-23} and 10^{-17} Ws^2 cm^{-3}. It turned out that the degree of polarization of the fluorescence radiation is strongly influenced both by hyperfine- and Zeeman-pumping of ground state sublevels and the finite length of the exciting laser pulse.

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2. Experimental

2.1. Fluorescence Cell

In designing the experimental set up (Fig. 1) the usual conditions for fluorescence light studies wer met [5]. The rectangular resonance cell (dimensions

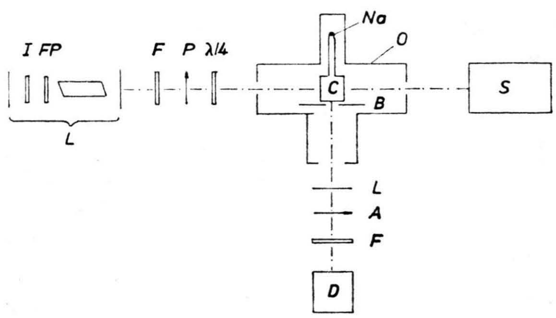

Fig. 1. Schematic diagram of the experimental arrangement. L dye laser; I interference filter; FP Fabry-Perot-etalon; F neutral filter; P, A linear polarizer and analyzer, respectively; $\lambda/4$ circular polaryzer; C fluorescence cell; B diaphragm; O oven; M grating spectrograph; D photoelectric detection chain, consisting of photomultiplier, integration amplifier, peak hold meter and data punch.

$10 \times 10 \times 30$ mm) made of fused silica was placed into an oven. Into a sidearm of the cell a Na mirror was distilled under high vacuum conditions. The temperatures of both the oven and Na mirror were kept constant within 0.2 °C. In order to prevent deposition of Na onto the cell walls, the oven temperature was held by about 10 °C above the mirror temperature. Temperatures were measured by means of Cu-Konstantan thermocouples. From the temperatures the Na densities were determined via the vapour pressure curve.

For reduction of stray light produced at the cell and oven walls a diaphragm was set in front of the cell fluorescence exit.

2. 2. Fluorescence Excitation

For the excitation of fluorescence a commercial flash lamp dye laser [6] was used with Rhodamin 6G as a dye. The energy of the laser light pulse was measured by means of a thermopile; typical values at 6000 Å without spectral narrowing were 10 mJ, corresponding to 10 kW during the pulse length of $\approx 1\ \mu s$ *.

Reduction of the spectral width of the laser emission was achieved by means of an interference filter combined with a Fabry-Perot etalon. The halfwidths were at most ≈ 0.05 Å *.

Tuning of the laser line frequency was achieved by tilting the filter (course tuning) and/or etalon (fine tuning) relative to the resonator axis. Course tuning it into the D_2 atomic resonance was effected by comparing visually its position relative to that of the D_2 line from a Na spectral lamp (not shown in the figure) on a glass plate in the focal plane of a grating spectrograph. Fine tuning was obtained by observing photoelectrically the maximum fluorescence intensity in the line center.

As linear polarizer and analyser polaroid sheets were used. For excitation with σ-light in addition a $\lambda/4$ mica plate was inserted into the exciting laser beam. The polarizing properties of sheets and plate under the action of the unweakened laser light beam were carefully tested. It turned out that they do not differ from those obtained with the light of a Na spectral lamp. In both cases it was found that to the σ-light leaving the circular polarizer an amount of $\approx 10\%$ π-light was admixed.

The intensity variation of the exciting beam over 5 orders of magnitude was achieved using Schott neutral transmission filters of known degree of transmission. The latter was measured by means of the laser beam actually used for fluorescence excitation.

2.3. Fluorescence Observation

Measurement of the fluorescence light pulses was performed photoelectrically using an EMI photomultiplier 9783/B. Time integration of each pulse was effected by means of an OEI operation amplifier with a time constant much larger than the pulse length. The integral value was digitally displayed on a peak hold meter and stored on a punch tape. Averages over 20 shots were then computed; standard deviations from the average values obtained were typically $\pm 2\%$.

Since the region of linearity of the peak hold meter reading covered only one order of magnitude, calibrated neutral filters were applied for reduction of the fluorescence radiation. By this method linearity between reading and fluorescence intensity could be established over the total range of investigation.

The statistical errors, together with the uncertainties introduced by using filters lead to a total error of the fluorescence intensity measurement of $\pm 20\%$.

The contribution of stray light from cell and oven walls to the fluorescence signal was measured by frequency shifting the laser line sufficiently away from the D_2 resonance. At small excitation intensities the stray light was negligibly small, at the highest intensities and smallest vapour pressures it amounted to $\approx 20\%$ of the fluorescence signal.

In order to avoid magnetic fields the measurements were performed during the heating free period (≈ 1 min) of the electrical oven heating. The earth magnetic field was not compensated for, since its depolarizing effect was considered to be negligibly small compared to the error bounds of the observed polarization values.

2.4. Polarization Degree

For the determination of the degree of linear polarization P the fluorescence signal was registered in the z- and x-position of the analyser. The registration in the two positions was performed alternatingly from pulse to pulse; thus possible long run shifts of the laser output power during 2×20 shots could be eliminated. Errors in P ranged between 3% at large and 15% at small excitation densities.

3. Results and their Interpretation

The interpretation of the data requires the knowledge of absolute values of the spectral density u_ν of the fluorescence exciting radiation produced by the laser beam inside the cell. From the energy E of the beam integrated over all angles and frequencies u_ν is obtained using the formula

$$u_\nu = E/F\, \Delta t\, c\, \Delta\nu$$

with F = laser beam crossection $= 0.2 \pm 0.04\ \text{cm}^2$, Δt = laser pulse length $\approx 1\ \mu s$, c = light velocity,

* quotation by the manufacturer CARL ZEISS, Oberkochen 1972.

$\Delta\nu$ spectral width of laser line $\approx 4 \times 10^9\ s^{-1}$. With a relative error of $\pm 10\%$ in the E-measurement, together with the uncertainties in the quotations for the values of $\Delta\nu$ and Δt, the overall error of u_ν may be estimated to about 50%.

3.1. Fluorescence Excitation Function

Figure 2: With increasing excitation density u_ν the fluorescence intensity I_F is observed to increase first and then to reach a constant value. This behaviour clearly reflects the dependence of the population of the $3\,^2P_{3/2}$ state on u_ν, which at sufficiently large u_ν saturates: the calculated relative number density $N_{3/2}/N_0$ of $^2P_{3/2}$ state atoms as function of u_ν coincides with the observed $I_F(u_\nu)$-function within the error limits.

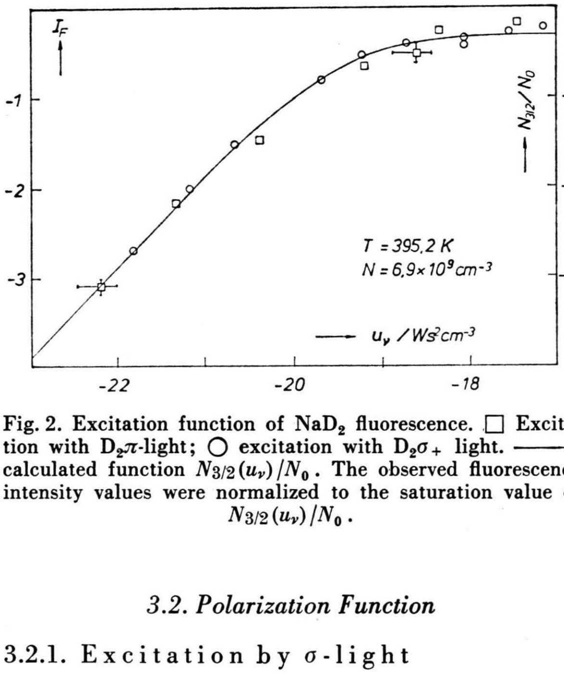

Fig. 2. Excitation function of NaD_2 fluorescence. □ Excitation with $D_2\pi$-light; ○ excitation with $D_2\sigma_+$ light. —— calculated function $N_{3/2}(u_\nu)/N_0$. The observed fluorescence intensity values were normalized to the saturation value of $N_{3/2}(u_\nu)/N_0$.

3.2. Polarization Function

3.2.1. Excitation by σ-light

Figure 3: At sufficiently small u_ν the observed polarization degree P remains constant. At larger u_ν it starts to increase and, for sufficiently small Na number densities N, saturates at very high u_ν. At given u_ν, P reduces with N; for the highest N the observed $P(u_\nu)$-function passes a maximum at very high u_ν.

The strong increase of P observed at intermediate u_ν may be interpreted in terms of optical pumping of the hyperfine- and Zeeman sublevels of the $3\,^2S_{1/2}$ state: From the hyperfine and Zeeman structure of

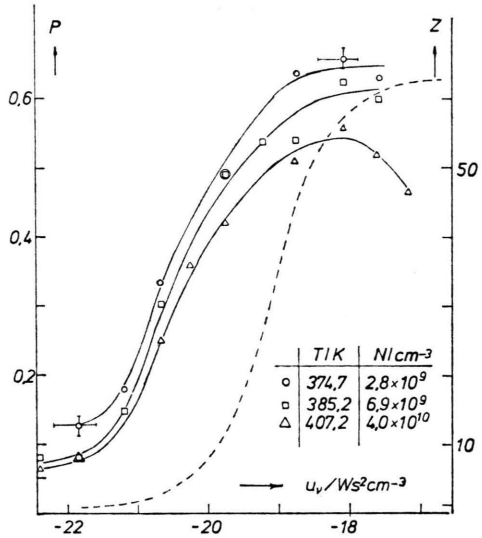

Fig. 3. Polarization functions of NaD_2 fluorescence. Excitation with $D_2\sigma_+$ light. —— observation; —·— calculated function $Z(u_\nu)$.

the transition $3\,^2S_{1/2} \rightarrow 3\,^2P_{3/2}$, together with the selection rules it follows, that the occupation of the $F = 2$, $M_F = +2$ substate of the $3\,^2S_{1/2}$ state and thus the degree of circular polarization of the fluorescence light will increase with u_ν.

A more quantitative explanation of the observed $P(u_\nu)$-function is possible, if in addition one takes into account the limited time of exposure of the atoms to the pump light due to the finite length of the exciting light pulse. For this purpose we estimate the number Z of pumping cycles during the pulse length Δt as function of u_ν. $Z(u_\nu)$ is roughly given by

$$Z(u_\nu) = \Delta t / [t_0(u_\nu) + \tau]$$

where τ = mean life time for spontaneous decay of the upper state, t_0 = mean life time of the ground state related to u_ν by

$$\frac{1}{t_0} = B\,u_\nu = \frac{1}{8\pi h}\,\lambda_0^3\,\frac{1}{\tau}\,u_\nu$$

where B is the Einstein absorption coefficient and λ_0 the wavelength of the transition. With $\tau = 1.6 \times 10^{-8}\ s$ ** and $\lambda_0 = 5890$ Å one obtains $B = 8.1 \times 10^{26}\ cm^3\ W^{-1}\ s^{-3}$.

Comparison of $Z(u_\nu)$ with the observed polarization function shows that P starts to increase if the number of pumping cycles during the pulse duration becomes larger than one. On the other hand, P

saturates at large u_ν, when Z no longer increases because stimulated emission does not contribute to pumping.

At small u_ν and small N the observed P-value of 0.12 ± 0.015 agrees well with the theoretical value $81/773 \approx 0.105$, obtained under the following assumptions: broad line excitation, equal population of all ground state Zeeman levels, no collisions. At large u_ν the measured P is considerably smaller then the theoretical value unity. This value would be expected after sufficiently long pumping, when all ground state atoms occupy the $F=2$, $M_F=+2$ level. It may be concluded, that the lower limit of the pumping time is comparably or larger than 10^{-6} s, the duration of the light pulse.

The observed reduction of P with increasing Na number density has been observed already by other authors [7–9]. It cannot been explained in terms of collisions since the crossections derived from the experimental data ($\approx 10^{-7}\,\mathrm{cm}^2$) are by several orders of magnitude larger than theoretical values ($\approx 10^{-11}\,\mathrm{cm}^2$). Instead, it has been proposed to interprete it as due to radiation diffusion [10]: Indeed, the optical depth $k_0 l$ of the fluorescent layer ($l \approx 1$ cm) in the line center is about 3×10^{-1} for the largest Na density ***, so that reabsorption is then no longer negligible.

On the other hand, the maximum in the $P(u_\nu)$-function, observed at the highest N, cannot easily be understood in terms of radiation diffusion, since this effect should in first order be independent of the radiation field density. More experimental data are clearly necessary before an analysis of this point can be offered.

3.2.2. Excitation by π-light

Figure 4: As in the case of σ-excitation one observes an increase of P with u_ν, however much smaller. This may again be understood in terms of pumping of ground state sublevels, which however should in the case of π-excitation be much less efficient, in agreement with observation.

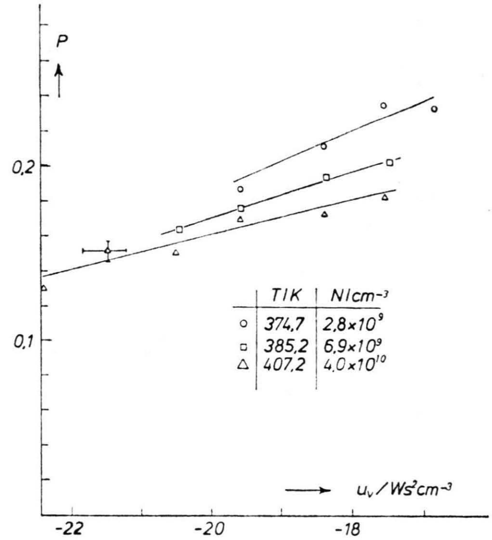

Fig. 4. Polarization functions of NaD_2 fluorescence. Excitation with $D_2\pi$ light.

For the smallest values of u_ν and N the observed polarization degree of 0.18 ± 0.01 agrees well with the theoretical value $81/427 \approx 0.19$. As with σ-excitation, P is observed to decrease with increasing N, in agreement with the findings of other authors [7–9]. Like there, the effect is supposed to be due to radiation diffusion.

4. Discussion

The proposed explanation of the observed effects in terms of optical pumping, although highly plausible, has still to be subject to several tests. One of them would consist of measuring the time dependence of P during the duration of the exciting pulse. If pumping is present, P should increase with the time elapsed after switching on the pulse. The question, whether the deviation of P from unity, observed in case of σ-excitation, is due to interruption of the pumping process, may be answered by a steady state experiment.

** value from Mitchell, Zemansky, l.c.

*** value obtained from $k_0 = 2\,N(\lambda_0/\Delta\lambda_D)\,r_0\,f$, with r_0 classical electron radius, $\Delta\lambda_D$ = Doppler width of the NaD_2 line ≈ 0.02 Å, f = oscillator strength = 0.7.

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