Stark-effect investigations of the sodium D_2 line

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The Stark effect of the sodium D_2 line was investigated by use of high-resolution laser-atomic-beam spectroscopy. The shift and splitting of the hyperfine components of the line could be obtained and compared with theoretical calculations. This allows the determination of the scalar and tensor polarizabilities with high accuracy to be 49.28(15) and $-21.97(10)$ kHz/(kV/cm)^2, respectively. These values are compared with semiempirical calculated polarizabilities. Additionally, the behavior of the relative intensity of the components could be studied. In fields greater than 100 kV/cm, the two groups of components with $M_J=\frac{1}{2}$ and $\frac{3}{2}$ are widely separated and the pattern of each group is independent from the field strength, showing an "electrical" Paschen-Back effect. The relative intensities agree satisfactorily with the computations.

I. INTRODUCTION

Shortly after Stark's discovery in 1913 of the splitting of the Balmer lines of hydrogen in an electric field, some physicists were interested in the Stark effect of the sodium D lines. One of the first was Ladenburg,[1] who published in 1914 his first work on the Stark effect of the D lines in which he stated that he observed no influence of the field (up to 46 kV/cm). In 1921 (Ref. 2) and 1924 (Ref. 3) he could present the first quantitative measurements and showed a quadratic dependence of the shift upon the field strength. In 1941, Jenckel and Kopfermann,[4] Kopfermann and Otzen,[5] and—independently of these authors—Gabler[6] were the first who used an atomic beam as absorbing medium and a Fabry-Perot interferometer to investigate small Stark shifts of resonance lines on calcium (Ref. 4), strontium (Ref. 5), and sodium (Ref. 6). In 1943, the work on sodium was continued by Kopfermann and Paul[7] and Gabler.[8,9] In these papers the hyperfine splitting of the ground-state level could be resolved and values of the scalar and tensor polarizabilities of the D_1 and the D_1 line were given.

After these measurements no new determination of the polarizabilities was done for about 30 years. Then, in 1970, Zimmerman[10] published investigations of the hyperfine structure of the $3\,^2P_{3/2}$ level in electric fields by use of the level-crossing method and Duong and Picqué[11] by means of the atomic-beam method.

Developments in the construction of tunable lasers made it possible to resolve completely the hyperfine structure of the sodium resonance lines and to restart investigations of the Stark effect of some levels of the sodium atom. Due to the specific technique of laser spectroscopy in presence of electric fields mainly highly excited states were investigated, e.g., by Littman *et al.*,[12] Gallagher *et al.*,[13] and Jeys *et al.*[14] Stark shifts of the resonance line were determined only by Hannaford *et al.*[15] using an optical transient method.

In 1982, we began with investigations of the sodium resonance lines under influence of external fields by means of laser-atomic-beam techniques, at first applying electric,[16] then magnetic,[17,18] and at last combined fields, taking advantage from experience in producing very high electric field strengths. Up to now some of the results on the D_2 line in electric fields were only published as small conference abstracts.[19,20]

Calculations of the polarizabilities and theoretical studies of the Stark effect in the hyperfine structure have been made by us and Murkawa and Yamamoto,[21] Khadjavi *et al.*,[22] Schmieder *et al.*,[23] Adelman and Szabo,[24] Schmieder,[25] and Duong and Picqué.[11]

II. BASIC CONCEPTIONS

Using an actively stabilized cw dye ring laser and a properly collimated atomic beam, the hyperfine components of the sodium D_1 and D_2 line can be resolved with a full width at half maximum (FWHM) near the natural linewidth. The laser absorption due to transitions between the sublevels of the lower and the upper level of the line can be detected by looking at the fluorescence light emitted by the excited atoms. This method was used by our group to investigate resonance lines of alkali-metal atoms in external fields. This paper reports experiments on the sodium D_2 line in high electric fields.

The former determinations of the polarizabilities[7–9] suggest to expect only small shifts. Therefore we had to use a field assembly which allows the application of very high electric fields. Nevertheless, the field strength should be known very accurately for a reliable determination of the polarizabilities. As can be learned, e.g., from Schmieder *et al.*[23] each hyperfine sublevel of $3\,^2P_{3/2}$ splits into $F+1$ components and shows an additional shift. The sublevels of the ground level, $3\,^2S_{1/2}$, show a shift only. With increasing field strength the line components are more and more mixed because the angular momentum of the atomic nucleus I is more and more decoupled from the total angular momentum J. Therefore a comparison of the experimentally observed pat-

terns with quantum-mechanical calculations is necessary to determine the polarizabilities of the line.

III. EXPERIMENT

The experimental arrangement was similar to those used for a high-precision measurement of the Stark shift of the sodium D_1 line:[16] The laser-induced fluorescence of an atomic beam (collimated approximately 1:400) was detected at two crossings of the atomic and the laser beam. One crossing was between the electric field plates, the second in a field-free area acting as a reference for the determination of the shift and the relative intensity of the Stark pattern. Each fluorescence area was imaged by use of a lens onto a light pipe and led to a photomultiplier. In our case of resonance fluorescence of course the fluorescence light has the same wavelength as the exciting laser light. Therefore stray light coming from the field electrodes would be detected. To suppress this stray intensity the atomic beam was chopped and the multiplier signals were amplified by means of lock-in techniques.

For field strengths up to 100 kV, the field assembly consists of two optical plane, silver-coated plates acting as a Fabry-Perot interferometer. Therefore an exact adjustment of the plates parallel to each other could be achieved. Using the dye laser and a lambdameter, the free spectral range of this Fabry-Perot interferometer could be measured very accurately. The field strength could then be calculated from the spacing d of the plates (0.55–0.8 mm; determined from the free spectral range) and the field voltage U, measured by means of a high-precision voltage divider. To make this technique of field-strength determination possible, the field-plate Fabry-Perot interferometer had to be actively stabilized by locking its spacing to a He-Ne laser. A detailed description of the field assembly used and the field-strength determination can be found in Refs. 26, 16, 27, and 28.

In higher fields, the silver coating of the plates can be destroyed by sparks between the electrodes. Therefore the use of stainless steel field plates was necessary, replacing the coated glass plates in the field assembly. A careful adjustment was necessary to avoid a broadening of the recorded line components due to an inhomogeneous electric field. The spacing ($\sim$0.5 mm) was determined by comparing the observed field pattern with another field pattern recorded in a precisely determined electric field (at $\sim$100 kV/cm). With this arrangement, fields up to 300 kV/cm could be reached.

IV. EVALUATION OF THE RECORDINGS

By help of a strip-chart recorder, four signals were recorded simultaneously while scanning the laser frequency. The first signal is caused by the crossing of the atomic beam with the laser beam in the field area and shows the field pattern, the second one comes from the field-free crossing. To avoid frequency offsets between these two signals, the laser beams were adjusted to give rise to their maximum fluorescence signal (at field voltage zero) for the same laser frequency. The third signal comes from a 1.5-m-long passively stabilized confocal

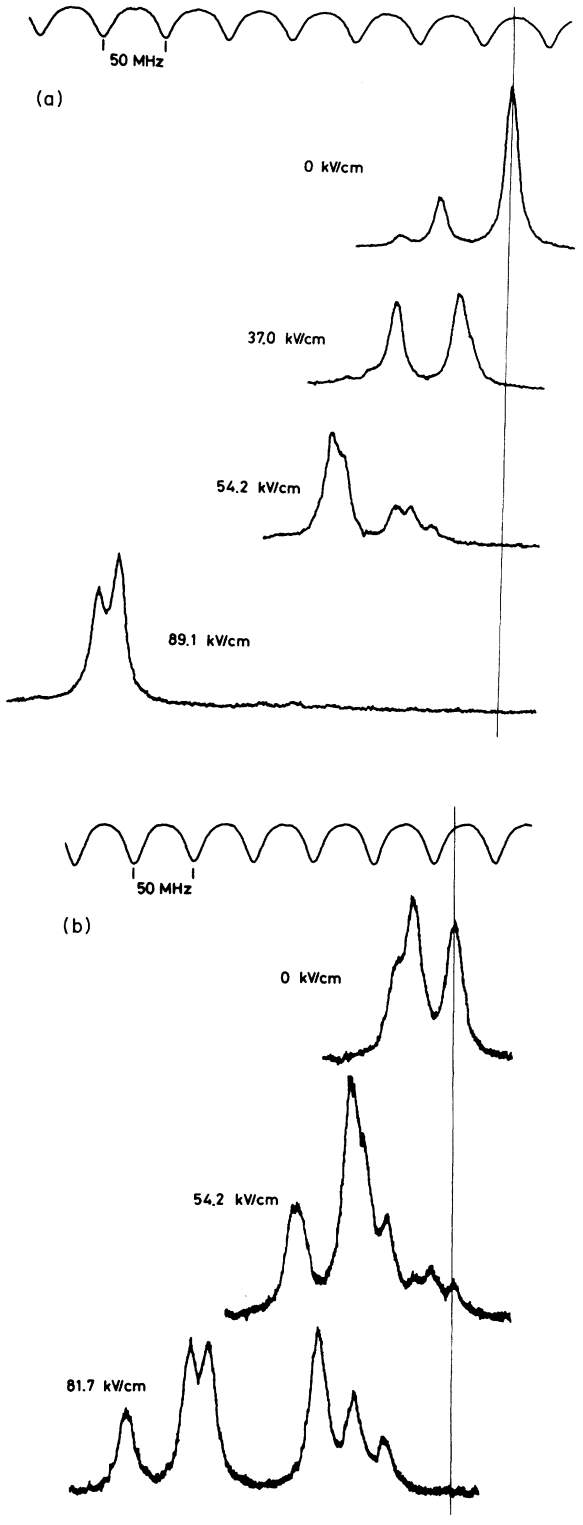

FIG. 1. Typical registrations of the hyperfine structure of the sodium D_2 line for different field strengths. (a) Low-frequency group ($F'=2\rightarrow F=3,2,1$) with π-polarized exciting laser light and (b) high-frequency group ($F'=1\rightarrow F=2,1,0$) with σ-polarized exciting laser light. The intensity of the (b) group is smaller; therefore the signal-to-noise ratio is not as good as in (a).

Fabry-Perot interferometer [free spectral range (FSR) 49.4(1) MHz]. The last signal comes from a second confocal cavity with 367-MHz free spectral range or—in a further series of recordings—from a high-finesse, actively temperature-stabilized confocal Fabry-Perot cavity with 375 mm length [FSR 197.50(8) MHz]. Either the 50-MHz or the 200-MHz cavity was used as a marker étalon. Their FSR was determined from the accurately known hyperfine structure of the sodium $3\,^2S_{1/2}$ level. The upper level of the D_2 line, $3\,^2P_{3/2}$, consists of four hyperfine levels $F=3$, 2, 1, and 0 separated from each other by approximately 59, 34, and 15 MHz, respectively. The ground-state level, $3\,^2S_{1/2}$, consists of two components $F'=2$ and 1 with a frequency difference of 1772 MHz. Due to this great splitting, the transition groups from $F'=2$ and $F'=1$ were registered separately, each for σ- and π-polarized laser light. Figures 1(a) and 1(b) show typical recordings.

A computer program was used to determine three parameters for each of the partly overlayed components, assuming Lorentzian line profiles: the frequency shift, the FWHM, and the relative intensity. At first, the recordings were digitized and the frequency of each digitized point was calculated by means of the trace of the 50-MHz (or the 200-MHz) étalon. Then estimated values for the site and the height of the components were fed into the computer. The following procedure calculated finally the parameters.

Let the observed pattern consist of n components. The first step was to subtract from the experimental pattern a sum of Lorentzian profiles (calculated by use of the estimated values), consisting of $n-1$ components. Starting now a least-square fit for the one component left (it should be the strongest) the estimated values for this component can be corrected (only corrections of start parameters can be calculated because the Lorentzian function led to nonlinear error equations which must be linearized). After this first correction another component (the next strongest) can be calculated and so on. Because the correction of a certain component has an influence on all previously calculated components the fit must be repeated as long as there are no more relevant changes in even one of all the $3n$ parameters.

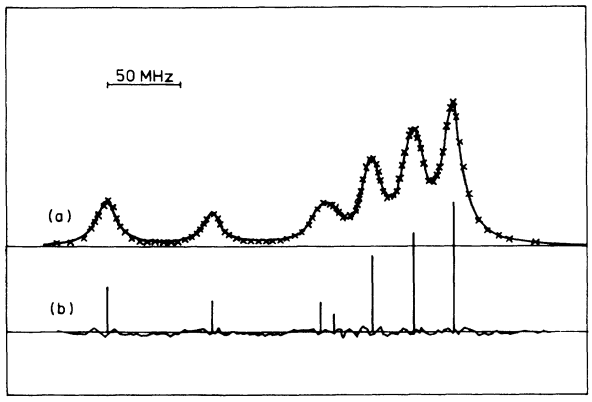

FIG. 2. Digitized registration (×) with fitted curve (a). In (b) the frequency and the height of the calculated components are shown, together with the deviation between digitized points and the fit (magnified 2 times).

For each recording, this fit procedure was done for the Stark pattern as well as for the field-free hyperfine pattern. Then all frequencies were referred to a certain field-free component. With this method the experimental signals can be fitted satisfactorily as can be seen in Fig. 2. Plotting the difference between experimental points and the fitted curve against the frequency, small systematic errors can be found in the line wings. When fitting a single component, the deviations are symmetrically around the line center. Therefore one can assume that the fitted values for the frequencies are correct.

V. CALCULATIONS

The Stark effect can be seen as the interaction of an uniform electric field with the atomic levels.[22,25] The interaction is given by the operator

$$H_E=-\mathbf{E}\cdot\mathbf{p}\ , \tag{1}$$

where $\mathbf{E}$ is the electric field and $\mathbf{p}=-e\mathbf{r}$ is the electric dipole moment operator. Since $\mathbf{r}$ has nonzero matrix elements only between states of opposite parity, the average value of H_E in any parity eigenstates vanishes. Hence the change in energy of a state $|a\rangle$ is given by the second-order perturbation formula

$$\Delta E(a)=\sum_b \frac{\langle a|H_E|b\rangle\langle b|H_E|a\rangle}{E_a-E_b}=\langle a|H'_E|a\rangle\ , \tag{2}$$

where the sum extends over all intermediate states $|b\rangle$ of the atom. Using the effective Hamiltonian H'_E, the Stark shift formally is equivalent to a first-order perturbation.

Taking the electric field direction as the z axis, the interaction Hamiltonian H'_E is given by

$$H'_E=-\tfrac{1}{2}\alpha_0 E^2-\tfrac{1}{2}\alpha_2\frac{3J_z^2-J(J+1)}{J(2J-1)}\ , \tag{3}$$

with the scalar polarizability

$$\alpha_0(\gamma,J)=-\frac{3}{2}\frac{1}{(2J+1)}\sum_{\gamma',J'}\frac{|\langle\gamma,J\|p\|\gamma',J'\rangle|^2}{E(\gamma,J)-E(\gamma',J')} \tag{4}$$

and the tensor polarizability

$$\alpha_2(\gamma,J)=2\left[\frac{10J(2J-1)}{3(2J+1)(J+1)(2J+1)}\right]^{1/2}\times\sum_{\gamma',J'}\frac{|\langle\gamma,J\|p\|\gamma',J'\rangle|^2}{E(\gamma,J)-E(\gamma',J')}(-1)^{J+J'+1}\times\begin{Bmatrix}J & J' & 1\\ 1 & 2 & J\end{Bmatrix}\ . \tag{5}$$

The Stark shift and splitting for an atom with hyperfine structure are obtained by diagonalizing the Hamiltonian

$$H=H_{\rm HFS}+H'_E\ , \tag{6}$$

where the hyperfine interaction is expressed in terms of

the magnetic dipole and electric quadrupole coupling constants A and B

$$H_{\mathrm{HFS}}=hA\mathbf{I}\cdot\mathbf{J}+hB\frac{3(\mathbf{I}\cdot\mathbf{J})^2+\frac{3}{2}\mathbf{I}\cdot\mathbf{J}-I(I+1)J(J+1)}{2I(I-1)J(2J-1)} . \tag{7}$$

The calculations for the sodium D_2 line were performed for given values of α_0 and α_2 by an improved version[18] of the computer program LEVEL, the original version created by Happer.[29] After replacing the magnetic perturbation given in[18] by the perturbation due to the electric field (3), the Hamiltonian (6) was diagonalized for each field strength. Its eigenvalues give the energy shift of the Stark levels. Figure 3 shows the calculated behavior of the upper level of the D_2 line, using $A=18.7$ MHz, $B=2.9$ MHz,[30] $\alpha_0=48.99$ kHz/(kV/cm) (Ref. 16), and $\alpha_2=-22$ kHz/(kV/cm)^2 (Ref. 31). As can be seen, some components are running very close together (in frequency distances smaller than the natural linewidth) and we cannot expect to resolve such components experimentally.

Since the tensor polarizability of the ground level is zero, the hyperfine levels of the ground state are only shifted but not splitted in the field and the splitting of the line itself reflects the splitting of the upper level. Therefore in Fig. 3 we used a value for α_0 which was measured as the difference between the scalar polarizabilities of upper and lower level of the D_1 line[16] in order to get immediately the shift and splitting for the two groups (due to $F'=1$ or 2) of the D_2 line.

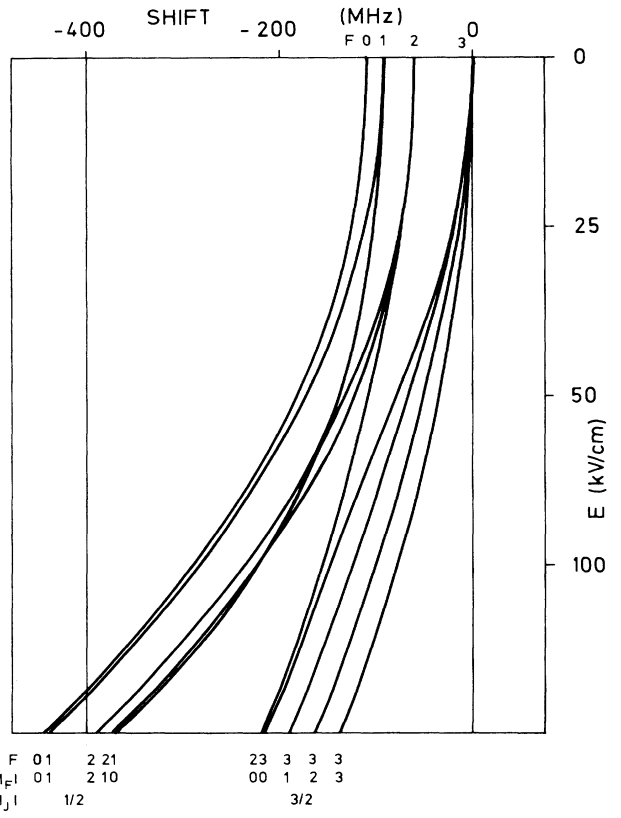

FIG. 3. Calculated Stark effect of the component groups of the sodium D_2 line. The splitting is due to the upper level ($3\,^2P_{3/2}$), the common shift is due to the difference between the shifts of upper and lower level ($3\,^2S_{1/2}$).

Using the computation method described in[18] we were able to calculate also the relative intensity of the Stark components $I,J',M_F'\rightarrow I,J,M_F$ where $M_F=M_I+M_J$.[32] Figures 4(a) and 4(b) show the results for the relative intensities of the Stark components. As can be seen, in fields higher than 120 kV/cm very small changes in the intensities occur. This indicates—in agreement with the splitting picture—that the coupling of I and J is destroyed at these field strengths.

The term $|\langle\gamma,J\|p\|\gamma'J'\rangle|^2$ in Eqs. (4) and (5) is equivalent to the so-called line strength

$$S(\gamma,J;\gamma',J')=|\langle\gamma,J\|p\|\gamma',J'\rangle|^2 . \tag{8}$$

Therefore, we were able to calculate values for α_0 and α_2 in a semiempirical way using tabulated line strengths of electric dipole transitions between the terms γ,J and γ',J' of energy $E(\gamma,J)$.[33–35] The results of this calculation together with those of other authors are shown in Table I.

VI. RESULTS

In Figs. 5(a) to 5(d) the observed Stark pattern for σ- and π-polarized exciting laser light are shown for field strengths up to 100 kV/cm, standardized to a common relative intensity by means of the reference track on the recordings (in some cases when a shifted Stark component has the same frequency as a certain reference component, optical-pumping effects must be taken into account). Under the peaks the frequency and the relative intensity due to theoretical calculations are shown. To interpret the observed pattern, one has to consider the transition rules either without or with field.

Without field, the transition rule $\Delta F=0,\pm1$, $\Delta M_F=0,\pm1$ is valid. Therefore the higher-frequency group of transitions consists of 3 components $F'=1\rightarrow F=2,1,0$; the group with the lower frequency has the quantum numbers $F'=2\rightarrow F=3,2,1$.

TABLE I. Calculated values for scalar and tensor polarizabilities for the levels of the sodium D lines [in kHz/(kV/cm)^2].

Level	Reference	α_0	α_2
$3\,^2P_{3/2}$	This work	85.5	−19.9
	11	86.2	−20.30
	21	84	−20
	23	86	−21
$3\,^2P_{1/2}$	This work	85.3	
	11	86.2	
	21	84	
$3\,^2S_{1/2}$	This work	41.0	
	11	39.0	
	21	40	
	24	40.0	

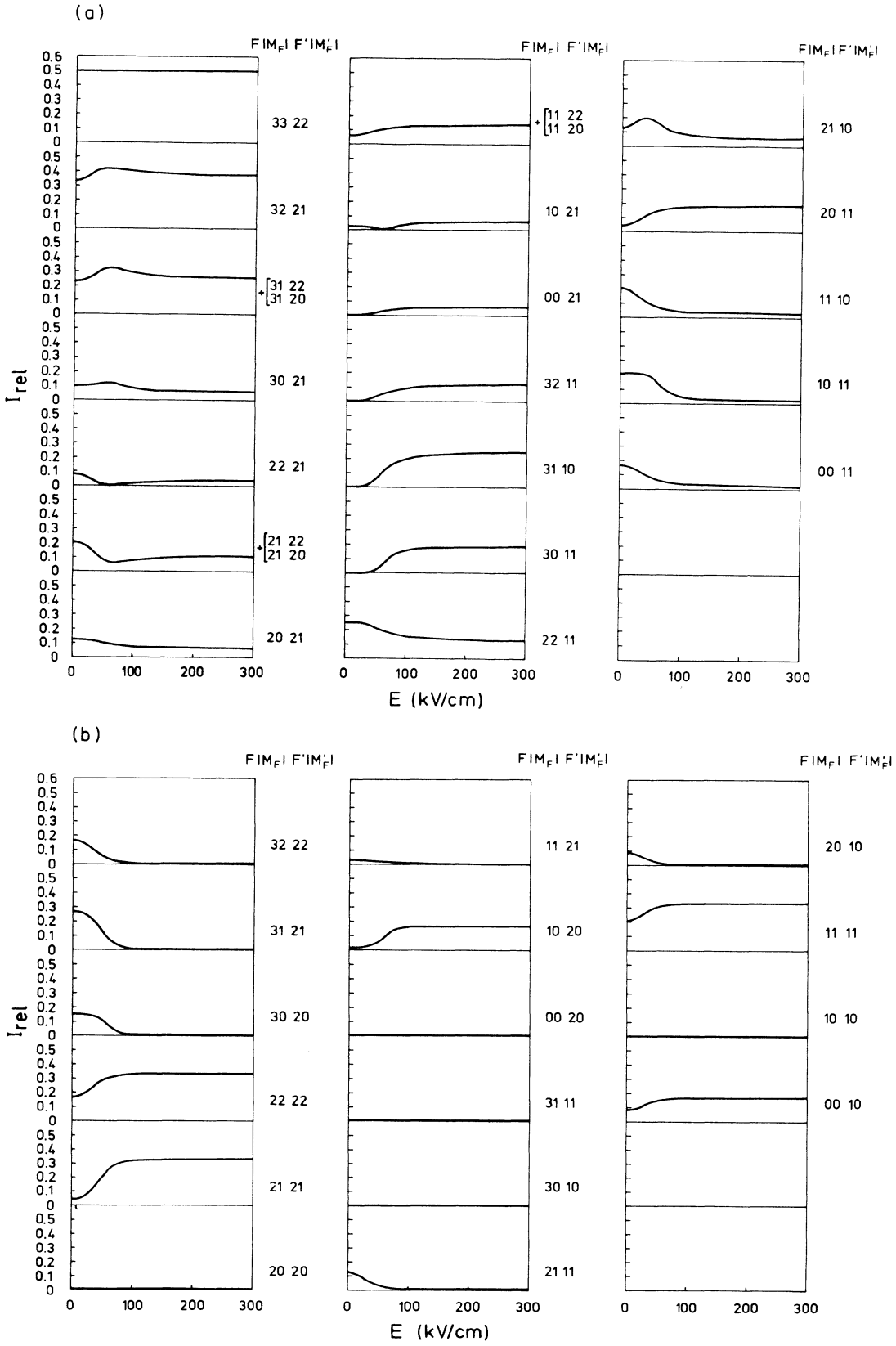

FIG. 4. (a) Relative intensities of the σ components of the D_2 line. Because of the hyperfine components of the lower level do not split up, some transitions led to components with the same frequency shift. For these cases, the relative intensities are added. (b) Relative intensities of the π components of the D_2 line.

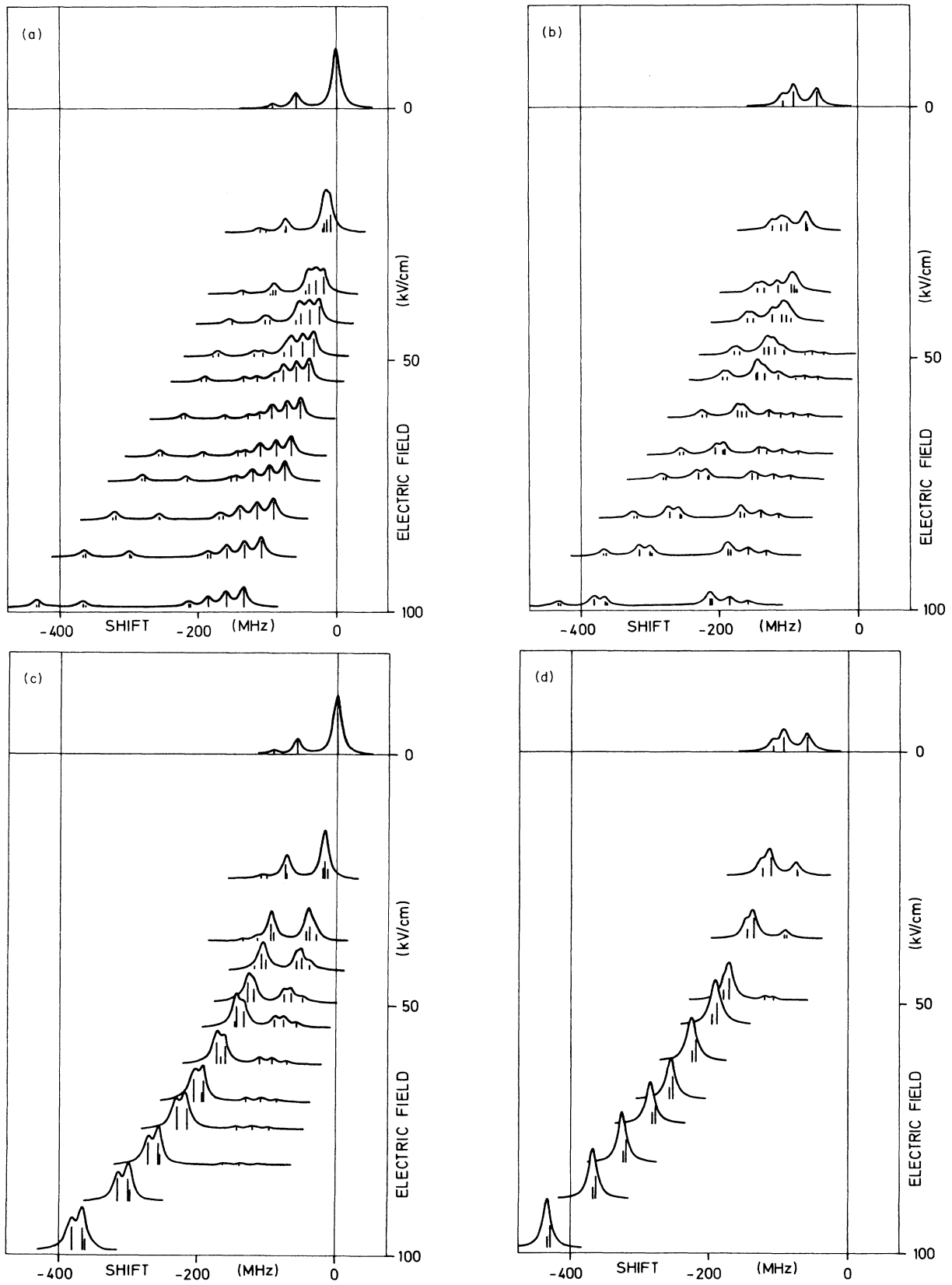

FIG. 5. Experimentally observed Stark pattern and calculated frequencies and relative intensities with σ-polarized exciting laser light for transition groups (a) $F'=2\rightarrow F=3,2,1$; (b) $F'=1\rightarrow F=2,1,0$; and with π-polarized exciting laser light for transition groups (c) $F'=2\rightarrow F=3,2,1$; and (d) $F'=1\rightarrow F=2,1,0$.

TABLE II. Experimental polarizabilities for the sodium D lines [in kHz/(kV/cm)^2].

Reference	α_0	α_2	$\alpha_0-\alpha_2$ ($\lvert M_J\rvert=\frac{1}{2}$)	$\alpha_0+\alpha_2$ ($\lvert M_J\rvert=\frac{3}{2}$)	$\alpha_0(D_1)$
This work	49.27(15)	−21.97(10)	71.24(18)	27.30(18)	
2			135(27)		
3			170(30)		
7			65.92(130)	25.19(90)	46.75(120)
8			67.2(?)	25.2(?)	45.6(?)
9			68.05(340)	24.52(120)	45.60(230)
10		−21.6(24)			
11			79.0(68)	29.4(34)	51.2(44)
15	47.2(33)	−28.1(39)			47.2(33)
16					49.99(11)

In low fields for σ- and π-polarized exciting laser light the recorded pattern are nearly the same as in the field-free case but shifted to lower frequencies. First small deviations in the shape of the pattern can be found at 25 kV/cm. With increasing field, an intermediate effect occurs decoupling I and J step by step. This decoupling is completed at approximately 100 kV/cm where the components are clearly separated in two groups with $\lvert M_J\rvert=\frac{1}{2}$ and $\frac{3}{2}$. Here $\lvert M_F\rvert=\lvert M_J+M_I\rvert$ and the selection rule $\Delta M_J=0$ (π polarization) and ±1 (σ-polarization) and $\Delta M_I=0$ is valid (strong field).

For the ground level, only a value of $M_J'=\frac{1}{2}$ is possible. For π polarization, only transitions $\lvert M_J'\rvert=\frac{1}{2}\rightarrow\lvert M_J\rvert=\frac{1}{2}$ will be allowed and all components in the Stark pattern belonging to $\lvert M_J\rvert=\frac{3}{2}$ should lose their intensity. This behavior can be seen in Figs. 5(c) and 5(d). On the other hand, in the case of the transition $F'=1\rightarrow F=2,1,0$ with σ-polarized excitation, the transition to $F=3$ is forbidden without field but the transitions $\lvert M_J'\rvert=\frac{1}{2}\rightarrow\lvert M_J\rvert=\frac{3}{2}$ are allowed in strong fields. Due to this fact, all Stark components starting from $F=3$ become more and more intense with increasing field, as can be seen easily in Fig. 5(b).

The only one case in which all transitions (with the exception of $F'=2\rightarrow F=0$ without field) were allowed at all field strengths is the transition group $F'=2\rightarrow F=3,2,1$ in σ-polarized excitation, shown in Fig. 5(a).

In order to compare the theoretical results given in Figs. 4(a) and 4(b) directly with the results of the experiments, one has to consider the polarization of the exciting laser beam as well as the direction of the observation (relative to the field axis) and the polarization of the observed fluorescence light. Observing perpendicular to the field (as had to be done because we must look between the field plates), the intensity of the (σ- and π-polarized) fluorescence light is proportional to the excitation probability given in the figures. The comparison is difficult because not all components are experimentally resolved. Drawing the calculated intensities under the observed pattern, one can find satisfactory agreement. It should be mentioned that for the whole Fig. 5 the intensities of Fig. 4 were adapted to the experiment with only one common factor.

As an example for the high field measurements, Fig. 6 shows the Stark effect of the D_2 line ($F'=2,1\rightarrow F=3,2,1,0$) up to 300 kV/cm for σ polarization. Investigating the field strength dependence of the two groups upon $M_J=\frac{1}{2}$ and $\frac{3}{2}$, e.g., in a field strength range >100 kV/cm, one will find quadratic behavior as stated in the older measurements[7–9] in which the hyperfine components of the groups were not resolved.

Before evaluating the data we had to consider a possible change in the hyperfine splitting of the ground state. As can be learned from Mowat,[36] this change is very small (only 0.5 kHz at 300 kV/cm) and can be neglected.

To determine precise values for the polarizabilities, a

TABLE III. Experimental scalar and tensor polarizabilities [in kHz/(kV/cm)^2] for the levels of the sodium D lines and the obtained hyperfine constants A and B [in MHz].

Level	Reference	α_0	α_2	A	B
$3\,^2P_{3/2}$	This work	88.98(87)	−21.97(10)	18.52(15)	2.98(34)
	10		−21.6(24)		
	11	84.8(84)	−24.4(50)		
	15	86.9(33)	−28.1(39)		
	30			18.69(9)	2.90(21)
$3\,^2P_{1/2}$	11	84.8(84)			
	15	86.9(33)			
	16	88.69(81)			
$3\,^2S_{1/2}$	38	39.7(8)			

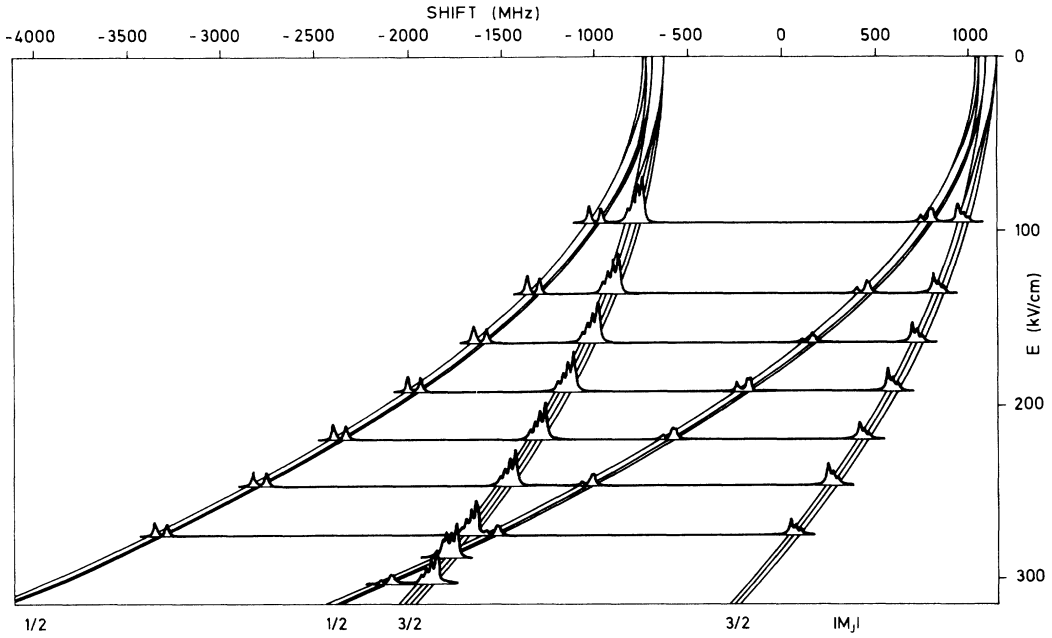

FIG. 6. Stark effect of the D_2 line in fields up to 300 kV/cm. At the highest field strength, the shift of the components with $|M_J|=\frac{1}{2}$ is more than 1.5 times the hyperfine splitting of the ground level and a crossing with components belonging to $|M_J|=\frac{3}{2}$ occurs.

fit procedure had to be made varying the polarizabilities and calculating the frequencies of the hyperfine components. This could be done by using a CERN program named MINUIT (Ref. 37) available at the "Rechenzentrum Graz." The program input was an array containing the frequencies of all components evaluated from our recordings. The columns of this data array were arranged by the quantum numbers of the components and each row corresponds to a certain field strength. Using the program, we fitted the scalar and tensor polarizabilities and simultaneously the hyperfine constants A and B of the upper level. Our experimental results together with values obtained by other authors are given in Table II. In Table III the polarizabilities of the levels are given using the best available experimental value for the polarizability of the ground state, $3\,^2S_{1/2}$.[38]

Comparing the value of α_0 [49.27(15) kHz/(kV/cm)^2] with that obtained for the D_1 line[16] [48.99(11) kHz/(kV/cm)^2], a small but significant difference in the magnitude of 0.5% can be stated. This difference in the polarizabilities led to our semiempirical calculations mentioned in Sec. V and could be confirmed by these taking into account the energy difference between $3\,^2P_{1/2}$ and $3\,^2P_{3/2}$.

In our errors in Table II there is included a systematical error due to the calibration of our marker étalons (0.2% or 0.1%), the systematical error of the field strength determination (0.03%, see e.g. Ref. 16) and a statistical error (standard deviation of the fit procedure, about 0.1%). In Table III the error of the polarizability of the ground state is added.

VII. CONCLUSION

Using laser-atomic-beam spectroscopy, the hyperfine structure of the sodium D_2 line could be studied in electric fields up to 300 kV/cm. From experimental data, a highly precise determination of the scalar and tensor polarizability of the line could be achieved. The pattern observed show very well the calculated relative intensities.

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