Stark effect of atomic sodium measured in a hollow cathode plasma by Doppler-free spectroscopy

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The Stark widths of the 5896-Å (3 2P 1/2 → 3 2S 1/2) neutral sodium hyperfine lines in the presence of charged perturbers were measured by means of laser saturation spectroscopy. The plasma containing the sodium atoms and the perturbers was generated in a hollow cathode discharge. The Baranger and Brechot theories for the line shape of neutral atom transitions in a plasma has been extended to hyperfine structure and applied to the hyperfine components of sodium D 1 line. The theoretical and experimental results were compared and the electronic density and temperature in the discharge were evaluated.

I. INTRODUCTION

Very high resolution spectroscopic techniques have been used by some experimentalists for studies on a hollow cathode discharge. The relatively high electron density (about 10^14 cm⁻³) obtainable by using this type of discharge in comparison with a conventional one (plane electrodes), without an excessive gas temperature, makes it possible to test experimentally the Stark effect on atomic levels of low energy.

Starting from the theory for Stark effect in plasmas developed by Baranger (1958) and Sahal-Brechot (1969) and with the help of some experimental results from the literature we have performed in Sec. IV a semiempirical study of the Stark broadening and shift of the D 1 line hyperfine components of atomic sodium in a plasma. The theoretical results have been tested experimentally in a hollow cathode negative glow, by means of the saturated absorption technique to avoid Doppler broadening. The experimental apparatus is described in Sec. II and the experimental values presented in Sec. III are finally discussed in Sec. V by comparison with the theoretical predictions. The results derived and some considerations about the discharge allows us to obtain information on the variation of the electronic temperature T e and density N e of the plasma generated.

II. EXPERIMENT

The experimental setup is shown in Fig. 1. An argon ion laser, model 171-09, was employed to pump a tunable dye laser, model 580 A, both from Spectra-Physics. The tunable laser was intensity stabilized, better than 1%, and the internal Fabry-Perot electronically locked to the cavity to avoid mode hopping. So, frequency jitter was minimized to ± 5 MHz.

With the help of an etalon 22.95 cm long, we have performed a calibration of the frequency axis. We shall only consider as representative those points which are separated by a frequency interval of 10 MHz, and the sweep linearity was estimated better than 3.8%.

The saturating beam was amplitude modulated and the signal coming from the probe beam fed into a lock-in amplifier. The demodulated and filtered signal was stored in a Hewlett-Packard 85-F computer through a digital voltmeter. The piezoelectric elements of the cavity are driven by a triangle-shaped variable voltage. This type of sweep has certain advantages such as the smooth return of the mirrors and consequently the possibility of performing averages of successive measurements in order to optimize the signal-to-noise ratio. The sweep time depends on the time constant (100 ms) of the detection system and on the number of representative points (~300) on the signal of frequency range swept; so the sweep time takes the value of 30 sec.

The discharge tube scheme is given in Fig. 2. Pyrex glass was used and the Brewster windows sealed with epoxy. The brass endcaps support the iron electrodes. The anode was an inverted T shape, and the cathode, which had 13-mm i.d. and 30-mm length is continuously refrigerated; a small portion of metallic sodium was located inside the cathode and distributed to the walls by the discharge.

The discharge was fed by an intensity-stabilized power supply through a ballast resistance (725 mΩ). The currents we have used lie between 100 mA and 200 mA requiring voltages ranging from 340 to 500 V. The discharge was initiated with helium gas at 500 mTorr, and the sodium was fed into the negative glow by sputtering, until a density depending on the discharge intensity is reached.

III. EXPERIMENTAL RESULTS

One of the spectra obtained for the D 1 line with the experimental setup described in Sec. II is shown in Fig. 3. The spectrum has a Gaussian background due to the velocity changing collisions suffered by the sodium atoms during their interaction with the laser beam. On the same figure we can see that the D 1 (i + ) line hyperfine components are resolved, but in the D 1 (i − ) line only the more intense component (3 2P 1/2, F = 2 → 3 2S 1/2, F = 1) appears while the other component (3 2P 1/2, F = 1 → 3 2S 1/2, F = 1) is very weak and it is masked by a crossover signal. On the other hand, the

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assymetry of the $D_1$ line background is due to the inverted
crossover signal that appears between these transitions.

From the theories recently developed by Berman, which take into account the effect of the velocity changing
collisions on the emitted line shape (in our case sodium $D_1$
lines), we were able to obtain the Stark widths of the transitions
under study. To do this, the theoretical signals were
fitted to the experimental ones. [The result of this fitting is
also shown in Fig. 3 (dotted line)]. From this fitting we
obtained the width of the Lorentzian part of the spectrum. To
obtain the contribution of the Stark effect we have subtracted
the contributions coming from the natural widths ($\sim 62$
MHz), power broadening ($\sim 40$ MHz), and
residual Doppler broadening due to the nonparallelism
between saturating and probe beams.

Figure 4 shows the dependence of the measured Stark
width on the discharge current in the range of interest. The
crossing hyperfine components have a similar behavior. The
contribution of mechanisms other than the Stark effect on
the signal has been tested in a saturated absorption
experiment in pure sodium.

IV. THEORY

We have calculated the effect of the electron and ion
collisions on the $D_1$ line hyperfine components of sodium
atoms with the help of the theories developed. Taking into
account the validity of the impact and classical path approximations, we have obtained the Stark broadening and shift of the transition between atomic hyperfine levels $|F_i\rangle$ and $|F_f\rangle$ of an alkali atom. It can be expressed in the isolated line approximation as follows:

$$
W + id = \frac{4\pi}{3} N_e \int_0^\infty f(u) du \sum_{F_i, f} |\langle F_i, f | r | F_i, f \rangle|^2 Q_{i, f} W^{(i)}_{i, f}
\times [a(Z_i, f_{\min}) + ib(Z_i, f_{\min})] + \frac{4\pi}{15}
\times N_e \int_0^\infty f(u) du \sum_{F_i, f} |\langle F_i, f | r | F_i, f \rangle|^2
\times Q_{i, f} W^{(2)}_{i, f} \omega_{i, f}^2 [a_q(Z_i, f_{\min}) + ib_q(Z_i, f_{\min})].
$$

In Eq. (1), we have taken $\hbar = m_e = a_0 = 1$, and
$$
Q_{i, f} = (2F_i, f + 1)(2J_i, f + 1)
\times (2J_i, f + 1)(2L_i, f + 1)(2L_i, f + 1),
$$
width has a value given by

$$
W_{sc} = \pi N_e \int_0^\infty v f(u) b_{min} du.
$$

The effect of the presence of ions has a negligible contribution in comparison with that of the electrons (about 1%) and as a consequence of this we can expect a Lorentzian
shape for the spectrum of every hyperfine transition of the $D_1$ line. The Stark width and shift are given by Eq. (1) plus the contribution due to strong collisions.

In order to test the validity of the approximations used to obtain Eq. (1), a previous diagnostic of plasma has been performed from the intensity ratio between the 4686-Å He-II line and the 5876-Å He-I line. We have estimated the values of $10^{14}$ cm$^{-3}$ for the electronic density and $7 \times 10^4$ K for the electronic temperature. With this value the impact approximation is justified because the collision frequency is

$$\nu \sim 10 \text{ MHz} < \tau_c^{-1} \sim 10^6 \text{ MHz},$$

$\tau_c$ being the collision time with an electron. The classical path approximation is also valid because the De Broglie wavelength associated with the electron $\lambda_e$ is less than the

![FIG. 2. Discharge tube scheme.](image)

![FIG. 3. Measured spectra of the $D_1$ line (continuous line) and theoretical fitting (broken line). Discharge current was 100 mA. (a) $D_1$ \((i-)\) component. (b) $D_1$ \((i+)\) component.](image)
average impact parameter
\[ b \sim 10^3 \text{ Å} \gg \lambda_e \sim 1 \text{ Å}. \]

For the \( D_1 \) line hyperfine components we can assume the isolated line approximation since the hyperfine splitting of \( 3^2P_{1/2} (192 \text{ MHz}) \) and \( 3^2S_{1/2} (1772 \text{ MHz}) \) are one and two orders of magnitude greater than the estimated Stark widths. In Fig. 5, the calculated Stark width is plotted versus the electronic temperature. \( (T_e \) and \( N_e \) being given in the range of interest). The evolution of the Stark width has been fitted to the function

\[ W(T_e, N_e) = \frac{N_e}{10^3} \left[ 85.24 - 63.2 \exp(-T_e/28.3 \times 10^3) \right], \tag{2} \]

where \( W \) must be expressed in MHz, \( N_e \) in \( \text{cm}^{-3} \), and \( T_e \) in \( ^\circ \text{K} \). This analytic form gives a good fitting of the theoretical evolution as it can be seen in Fig. 5 and it will be useful to study the dependence of \( N_e \) and \( T_e \) on the discharge current.

V. DISCUSSION

Experimentally it is observed that the Stark width and the voltage difference between the electrodes decreases when the current in the discharge increases. From this behavior we can conclude that an increase of the current produces a decrease of the electronic temperature.

On the other hand, the optical density measurement on the \( D_1 \) line gives for variation of the sodium density a dependence on the current intensity \( I \) of the discharge given by

\[ n_{Na}(I) = 3.6 \times 10^{10} \exp(0.0251), \]

where \( n_{Na} \) is given in \( \text{cm}^{-3} \) and \( I \) in mA.

The electronic temperature \( T_e \) can be expressed as

\[ T_e = T_g + \frac{T}{[\text{coll (He)}] + [\text{coll (Na)}]}, \]

\( T_g \) being the gas temperature. The second term informs on the energy excess of the electrons accelerated from the cathode walls into the negative glow by the electronic field of the dark space and braked by collisions with helium and sodium atoms. On the other hand, from the evolution of the Gaussian width of the background as a function of the intensity current, Fig. 6, we can extract the quadratic law for the gas temperature in \( ^\circ \text{K} \)

\[ T_g = 9.80 \times 10^{-4} (2.41 + 430)^2. \]

So, the expression for the electronic temperature \( T_e \) as a

FIG. 4. Measured Stark width and theoretical fitted evolution (broken line).

FIG. 5. Theoretical evolution of the Stark width and fitting to \( W(T_e, N_e) \) (continuous line).

FIG. 6. Evolution of the Gaussian background width vs the current discharge. Linear fitting (broken line).

FIG. 7. Calculated evolution of the electronic density \( N_e \) vs the discharge current intensity \( I \).

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DISCHARGE CURRENT (mA)

FIG. 8. Calculated evolution of the electronic temperature $T_e$ vs the discharge current intensity $I$.

The function of $I$ can be written as follows

$$T_e = 9.80 \times 10^{-4}(2.4I + 430)^2 + \frac{1}{B + C \exp(0.025I)},$$

where $B$ and $C$ are parameters related to the total collision cross sections between the electron and helium and sodium atoms, respectively.

The geometry of hollow cathode in our discharge tube allows us to write

$$N_e T_e^{1/2} = \alpha I,$$

$\alpha$ being a parameter related to the geometry and kinetic of electrons. From Eq. (3), Eq. (4), and the expression (2) is obtainable the theoretical evolution of $W(T_e, N_e)$ as a function of the current intensity, $I$. The fit of the expression obtained to the experimental results (plotted in Fig. 4) leads to the values of the parameters $B$, $C$, and $\alpha$. The substitution of these values into Eqs. (3) and (4) leads to the evolution of $N_e$ and $T_e$ on $I$. They are plotted in Figs. 7 and 8, respectively.

In these plots we can see that the electronic density increases from $3.05 \times 10^{13}$ cm$^{-3}$ at 100 mA to $3.69 \times 10^{14}$ cm$^{-3}$ at 200 mA; under the same conditions the electronic temperature decreases from $15 \times 10^4$ K to $2.3 \times 10^4$ K. The estimated error in the determination of $N_e$ is better than 12% and better than 20% in that of $T_e$.