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# Hyperfine Optical Pumping of Sodium Vapors*

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A new method has been designed for obtaining the hyperfine optical pumping of Na vapor. The pumping lamp is placed in a magnetic field and the light emitted in the field direction is used for excitation after crossing a circular polarizer selecting one of the Zeeman lines. This method has two interesting features: (i) The atoms can be pumped either in the lower or in the upper hyperfine sublevel by simply rotating a polarizer by an angle of $\frac{1}{2}\pi$; (ii) the hyperfine pumping is obtained using both the $D_1$ and $D_2$ resonance lines without filtering losses. The pumping efficiency is good and it has been possible to measure the spin-exchange cross section between sodium atoms by studying the hyperfine relaxation rates as a function of the sodium density. The result is $\sigma_{ex} = (1.109 \pm 0.005) \times 10^{-14}$ cm$^2$.

## I. INTRODUCTION

The ground state of the alkali metals is ${}^2S_{1/2}$ and is split into two hyperfine sublevels $F^+ = I + J$ and $F^- = I - J$, where $I$ is the nuclear spin. Then the resonance lines $D_1$ and $D_2$ are also split into two hyperfine components, since the hfs of the excited states ${}^2P_{1/2}$ and ${}^2P_{3/2}$ is much smaller. We can excite only the atoms that are in a given hyperfine sublevel by filtering the corresponding component of the resonance lines. When the excited atoms return to the ground state they fall also in the other sublevel where they accumulate. In this way we obtain the hyperfine optical pumping. The main problem is the filtering of one component of the resonance line, since the hyperfine splitting is about $10^{-1} \approx 10^{-2}$ Å. Filters are obtained by means of a cell containing the vapor of the alkali itself and using separated isotope or a magnetic field. In this way the hyperfine optical pumping of Rb[1] and Cs[2] has been obtained.

For Na and K there is another difficulty. The Doppler width of the lines is about equal to the hyperfine splitting. In the case of Na, the hyperfine splitting is 1.8 GHz and the Doppler width is 1.5 GHz. With this value an absorption filter, like that designed for Cs,[3] does not work well. In this paper we describe a new method for hyperfine pumping of the alkali atoms that overcomes this difficulty.[4]

Let us consider an alkali lamp placed between the poles of a magnet. In the field direction only the $\sigma^+$ and $\sigma^-$ lines are emitted, both circularly polarized but in opposite direction. The high-frequency line ($\sigma^+$) can be easily separated from the low-frequency line ($\sigma^-$) by means of a $\frac{1}{4}$ $\lambda$ plate and a polarizer. When the polarizer is turned by an angle of $\frac{1}{2}$ $\pi$, the transmitted line is the $\sigma^-$. For a given value of the magnetic field, the $\sigma$ lines are partially superimposed to the zero-field hyperfine components $F^+ \leftarrow F'$ and $F^- \leftarrow F'$ (Fig. 1). Then the light emitted by the lamp can be used for the hyperfine optical pumping. The $\sigma^+$ line pumps the atoms in the upper level $F^+$, and the $\sigma^-$ line in the lower $F^-$. Another advantage of this method is that both the $D_1$ and $D_2$ lines can be used simultaneously. Then there are no losses in preparing the pumping light beam. This method has been applied to the hyperfine optical pumping of Na vapor with good results.

## II. EXPERIMENTAL APPARATUS

The most important part of the experimental apparatus is the pumping lamp, since it must fulfill rather opposite conditions. It must be as bright as possible, and on the other hand the $\sigma$ lines must be well circularly polarized and as close as possible to the Doppler limit width. Self-reversal and related effects must be carefully avoided.[5] The shape and polarization of the lines was studied by means of a piezoelectric scanning Fabry-Perot interferometer. This experimental apparatus is shown in Fig. 2. The magnetic field is generated by means of an electromagnet, which was modified in order to in-

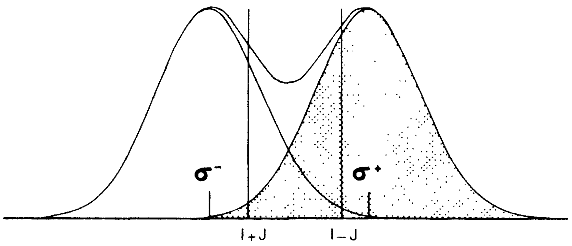

FIG. 1. Zeeman pattern in the field direction. The position of the hyperfine transitions is also shown.

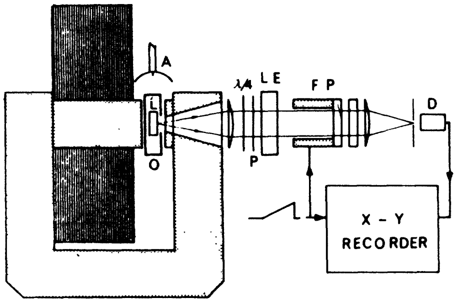

FIG. 2. Experimental apparatus for measuring the shape of the pumping lamp lines. A, microwave antenna; L, Lamp; O, oven; P, polarizer; LE, Lyot-Evans filter; FP, Fabry-Perot interferometer; D, photomultiplier.

crease the light output in the field direction. One coil was removed and the hole drilled in the pole was made conical with a useful solid angle $\Omega \approx 1$ sr.

Several types of lamp have been tested and the best results have been obtained with a microwave-excited electrodeless lamp. The lamp bulb of cylindrical shape, 2 cm in diameter and 1 cm in height, is made of a special Na resistant glass and has a very long life. The bulb is filled with about 1-Torr Ar and a few mg of Na metal and it is placed in a little oven between the poles of the magnet. We used for the excitation a dc-powered cw magnetron giving a maximum output of 100 W at 2.5 GHz. The intensity of the emitted light was stable within 0.1%.

The light emitted in the field direction crossed a $\frac{1}{4}\lambda$ plate, a linear polarizer rotable at angles of $+\frac{1}{4}\pi$, 0, and $-\frac{1}{4}\pi$ with respect to the axis of the $\frac{1}{4}\lambda$ wave plate, and a Lyot-Evans filter,[6] by means of which either the $D_1$ or the $D_2$ line can be filtered out. Then we measured for both lines the spectral distribution of the light emitted polarized $\sigma^+$ or $\sigma^-$ (linear polarizer at angles $+\frac{1}{4}\pi$ and $-\frac{1}{4}\pi$, respectively), or the sum $\frac{1}{2}(\sigma^+ + \sigma^-)$ (polarizer at angle 0) as a function of the magnetic field strength. The result for a field of 1.7 kG is given in Fig. 3. The filtering of the $\sigma^+$ or $\sigma^-$ lines seems to be incomplete for the $D_1$ line. This effect is due to the incomplete Back-Goudsmit effect. The same lines fulfilling the selection rule $\Delta m_F = \pm 1$ are still sufficiently intense to appear in the recorded spectrum for magnetic field below 2 kG.[5-7] In the $D_2$ line this effect is less observable because the hfs splitting of the upper level is smaller. Since the intensity of this unwanted line is greatly enhanced by the effect of self-reversal,[5] the temperature of the lamp was carefully monitored. The best results were obtained for an oven temperature of 170–175 °C. The highest hyperfine pumping efficiency was experimentally found for a magnetic field between 1 and 1.2 kG. The experimental apparatus for obtaining the hyperfine optical pumping of sodium is shown in Fig. 4. The pumping light beam, after crossing the circular polarizer, was focused on the shutter in order to decrease the beam diameter and thus decrease the effective closing time of the shutter. The light was then sent to the absorption cell and the fraction absorbed by the Na vapor was measured by means of a photomultiplier in order to detect the hyperfine pumping. The measurement of the relaxation time of the observable $\langle S \cdot I \rangle$ which is proportional to the differences of the occupation numbers of the two hyperfine sublevels, was carried out in two ways. The first was Franzen's method,[8] and here we used an inverted photographic shutter. A box camera shutter was used, instead, for the second method. The closing time was about 2 msec and when the shutter was closed, about 1% of the pumping light was still transmitted from a small hole drilled in the moving flag. The intensity of this light was not enough for obtaining any detectable hyperfine pumping, but it was sufficient for continously monitoring the transmis-

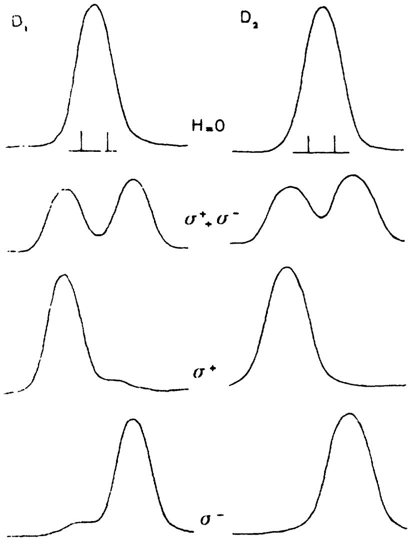

FIG. 3. Shape of the $D_1$ and $D_2$ lines for a magnetic field of 1.7 kG. Oven temperature is 170 °C. The theoretical position of the hyperfine lines in zero field is also shown.

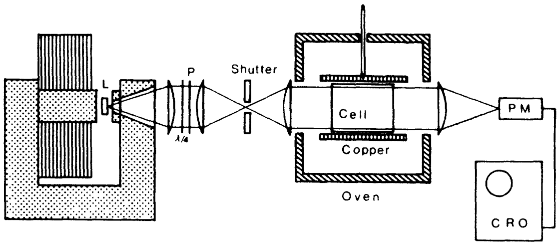

FIG. 4. Experimental apparatus for the hyperfine optical pumping of sodium vapor.

sion of the vapor. Since this signal is proportional to the population of the hyperfine sublevel evacuated by the pumping light, the relaxation of the quantity $\langle S\cdot I\rangle$ is directly recorded. Since the pumping light is linearly polarized after crossing the circular analyzer, the vapor can also be aligned. At any rate, this effect is much smaller than the hyperfine pumping[9] and depends upon the direction of the magnetic field on the absorption cell. For a suitable choice of the magnetic field direction, the alignment can be completely eliminated. With this method it was possible to employ the signal-averaging technique in order to increase the signal-to-noise ratio. In Fig. 5 a typical result is shown. For comparison the relaxation of the quantity $\langle S_z\rangle$ is also shown, i.e., the observable connected with the optical pumping with circularly polarized light. This signal was obtained by measuring the Faraday rotation of the oriented vapor. For this purpose we have used another experimental apparatus that has been described in Ref. 10. The absorption cells of cylindrical shape were made out of Pyrex glass with an outside diameter of 8 cm and a height of about 12 cm. The cell was placed in a thermostatically controlled oven and it was surrounded by a cylindrical copper sheet 3 mm thick in order to keep the temperature as uniform as possible. In this way the temperature of the cell was measured with an error of 0.3 °C. Great care was taken in the filling of the cells. Our aim was to measure the relaxation time of the quantity $\langle S\cdot I\rangle$ as a function of the Na vapor density. In order to obtain reproducible results it was necessary that the Na density be that of the saturated vapor. The Na metal was then distilled several times under vacuum and introduced into the cell in a quantity sufficiently large to neutralize the chemical reactivity of the cell walls. Moreover, any contamination due to diffusion pump oil or mercury backstreaming and to buffer-gas impurities was carefully avoided. Our vacuum apparatus was built with a molecular sieve as a forepump and a high-vacuum ionic pump. The buffer-gas pressure was measured by means of a Bourdon-type manometer.

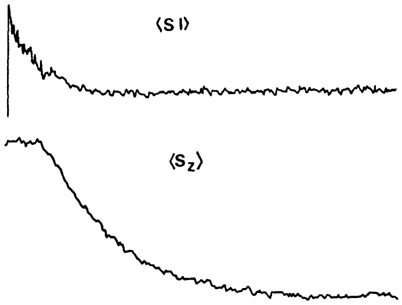

FIG. 5. Relaxation signal of a cell with 30 Torr Ne at 111 °C. The upper curve is the relaxation of the hyperfine pumping ($\tau_{\langle\vec{s}\cdot\vec{i}\rangle}=0.057$ sec), the lower curve is the relaxation of the Zeeman pumping ($\tau_{\langle\vec{s}_z\rangle}=0.245$ sec). Different experimental apparatus were used for the two measurements.

## III. EXPERIMENTAL RESULTS

### A. Measurement of Na Spin-Exchange Cross Section

The hyperfine optical pumping of Na vapor was observed in cells with He, Ne, or Ar as a buffer gas, and in the temperature range 105–135 °C. The relaxation time increases by increasing the buffer-gas pressure. However there is an upper limit to the pumping efficiency due to the resonance-line broadening which is proportional to the buffer-gas density. When the broadening is larger than the hyperfine splitting, the hyperfine pumping is no longer possible. Ne is the best buffer gas, since it has the smaller broadening constant.[11] With Ne, the hyperfine pumping was observed for a pressure up to 40 Torr, while for He and Ar the maximum pressure was 30 and 20 Torr, respectively. We can calculate the spin-exchange cross section between Na atoms from the measured value of the relaxation time of the quantity $\langle S\cdot I\rangle$. Since $\langle S\cdot I\rangle$ relaxes with a single expotential within experimental accuracy, we can put

$$1/\tau=1/\tau_0+1/\tau_{ex}\,, \tag{1}$$

where $\tau$ is the total observed relaxation time, $1/\tau_0$ is the relaxation probability due to collisions between Na atoms and the buffer-gas atoms or the cell walls, and $1/\tau_{ex}$ is the relaxation probability due to the spin-exchange cross section. $1/\tau_{ex}$ is proportional to the density of the Na atoms, while $\tau_0$ is practically independent of the temperature of the cell. A temperature dependence of $\tau_0$ appears at higher pressures of the buffer gas, and it is due to the diffusion process which prevents the uniform distribution of the pumped atoms inside the cell.[12] At low pressure and in the small temperature range scanned, however, this effect is negligible and $\tau_0$ can be considered as a constant. This has been directly tested by measuring also the relaxation time of the quantity $\langle S_z\rangle$, which is independent of the spin-exchange collision. The diffusion problem is identical for both $\langle S\cdot I\rangle$ and $\langle S_z\rangle$ in the same cell. Then, if the relaxation time of $\langle S_z\rangle$ is found to be temperature independent, $\tau_0$ is also temperature independent. The measures for a cell with 30 Torr Ne are given in Fig. 6 and similar results were obtained for all other cells. The spin-exchange cross section can be calculated by

$$\sigma_{ex}=\frac{\Delta\,(1/\tau)}{\Delta\,N}\,\frac{1}{V}\,, \tag{2}$$

where $V=4\,(RT/\pi\,M)^{1/2}$ is the mean relative velocity of the Na atoms, and $N$ is the number of Na

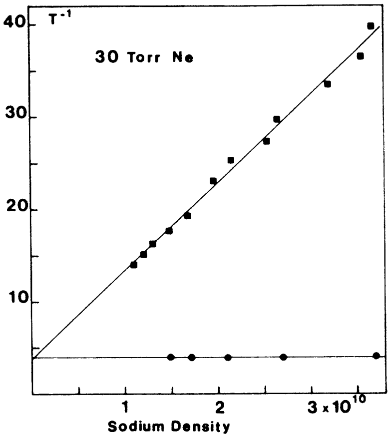

FIG. 6. Relaxation time in units of inverse seconds as a function of the Na vapor density, in atoms/cm$^3$. ■: hyperfine relaxation, $\langle S \cdot I \rangle$. ●: electronic spin relaxation $\langle S_z \rangle$. For this cell we found $\sigma_{ex} = 1.12 \times 10^{-14}$ cm$^2$.

atoms per cm$^3$. A plot of the measured values of $1/\tau$ versus $N$ is given in Fig. 7. Since the Na-vapor density was not known below the value of $10^{12}$ atoms/cm$^3$ (~180 °C), we measured it in the range 180–100 °C. The value of $N$ was calculated by measuring absolute value of the integral of the $D_1$ absorption line by means of magnetic scanning technique.[13] The measurements were carried out with several cells prepared with the same method used for the cell in the hyperfine pumping experiment. From the results we computed the following formula

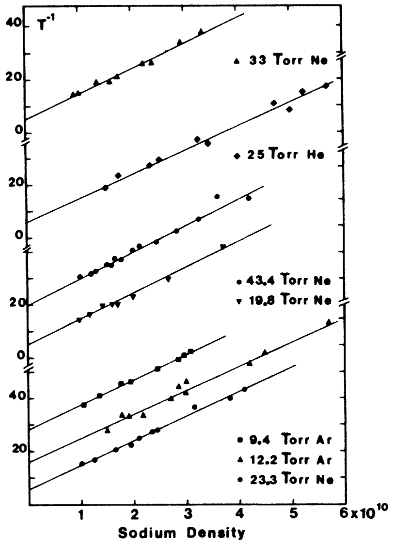

FIG. 7. Hyperfine relaxation time in units of inverse seconds as a function of Na vapor density, in atoms/cm$^3$. From the top we calculated the following values of $\sigma_{ex}$ in units of $10^{-14}$ cm$^2$: 1.11; 1.07; 1.17; 1.12; 1.12; 1.08; 1.08.

TABLE I. Comparison of the experimental and theoretical spin-exchange cross section of alkali atoms ($10^{-14}$ cm$^2$).

| Authors and Ref. | Na-Na | K-K | Rb-Rb | Cs-Cs | $T$(°K) |
|---|---|---|---|---|---|
| Experimental values | | | | | |
| Jarret[a] | | | 1.85 ± 0.23 | | 363 |
| Gibbs-Hull[b] | | | 1.9 ± 0.2 | | 351 |
| Grossetete[c] | | 2.7 ± 0.7 | 2.1 ± 0.3 | 2.20 ± 0.35 | 300<br>350 |
| Ernst-Strumia[d] | | | | 2.20 ± 0.15 | 300 |
| Moretti-Strumia[e] | 1.109 ± 0.005 | | | | 390 |
| Ressler-Sands-Stark[f] | 1.03 ± 0.21 | 1.45 ± 0.21 | 1.9 ± 0.2 | 2.06 ± 0.2 | 500<br>700 |
| Theoretical values | | | | | |
| Smirnov-Chibisov[g] | 0.98 | 1.41 | 1.57 | 1.80 | 500 |
| Dalgarno-Rudge[h] | 1.1 | 1.5 | 1.6 | 1.9 | 540 |
| | | 1.6 | 1.8 | 2.0 | 300 |
| Chang-Walker[i] | 1.24 | 2.44 | 2.70 | 2.82 | 300 |
| | 1.21 | 2.38 | 2.64 | 2.77 | 405 |

[a] Reference 13.
[b] H. M. Gibbs and R. J. Hull, Phys. Rev. 153, 132 (1967).
[c] F. Grossetete, D. Sc. dissertation, University of Paris, 1967 (unpublished).
[d] Reference 2.
[e] This work.
[f] Reference 15.
[g] B. M. Smirnov and M. I. Chibisov, Zh. Eksperim. i Teor. Fiz. 48, 939 (1965) [Soviet Phys. JETP 21, 624 (1965)].
[h] Reference 16.
[i] C. K. Chang and R. H. Walker, Phys. Rev. 178, 198 (1969).

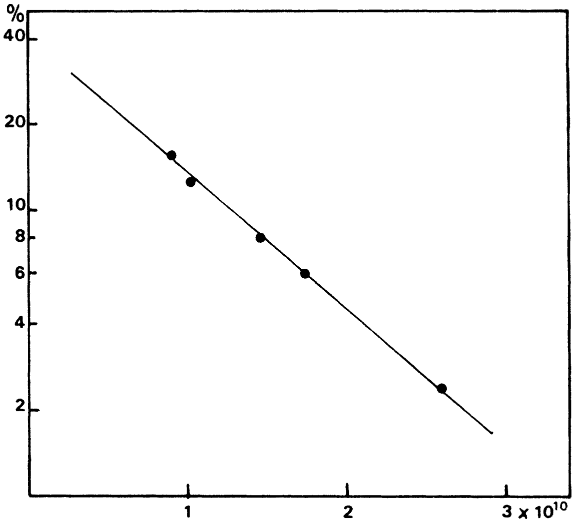

FIG. 8. Hyperfine pumping efficiency as a function of Na-vapor density and for a cell filled with 30-Torr Ne. The points are calculated by means of Eq. (5).

for the density of the saturated sodium vapor[14]:

$$\log_{10} N = A - B/T + C \log_{10} T, \tag{3}$$

where $A = -40.6658$, $B = 781.7616$, $C = 20.4594$, and the estimated error is 2–3%.

The value of $\sigma_{ex}$ was obtained for eight cells and the average value was found to be

$$\sigma_{ex} = (1.109 \pm 0.005) \times 10^{-14}\ \mathrm{cm}^2,$$

where the quoted error is twice the standard error. Our value is in good agreement with that obtained recently by Ressler, Sands, and Stark,[15] using an entirely different method.

In Table 1 we report the known values of the spin-exchange cross section of the alkali metals. We found that the theoretical values computed by Dalgarno and Rudge[16] give the best fit of the experimental results.

### B. Measurement of Pumping Efficiency.

The efficiency of the hyperfine optical pumping can be calculated by measuring the absorption of the cell, when the vapor is pumped ($A_p$) and when it is at the thermal equilibrium ($A_t$). The population of the hyperfine sublevels $F = 1$ and $F = 2$ are $n_1^t = \frac{3}{8}N$ and $n_2^t = \frac{5}{8}N$ when the vapor is at the thermal equilibrium. Now suppose that we pump the Na vapor from the $F = 1$ sublevel to the $F = 2$ sublevel. As a first approximation, the signal received by the photomultiplier is proportional to the absorption of the atoms in the $F = 1$ sublevel. Then we obtain

$$A_t = I_c - I_t = K n_1^t, \quad A_p = I_c - I_p = K n_1^p \tag{4}$$

where $K$ is a constant and $I_c$, $I_t$, $I_p$ are, respectively, the signals of the photomultiplier when the cell is cold (no Na vapor), at the thermal equilibrium, and with hyperfine pumping.

The fraction of atoms transferred from the $F = 1$ level to the $F = 2$ level is given by the formula

$$\Delta n_1/n_1^t = (A_t - A_p)/A_t\,. \tag{5}$$

In Fig. 8 this quantity is given as a function of the Na density for a cell filled with 30-Torr Ne. No significant difference was found between the pumping in the upper $F = 2$ level or in the lower $F = 1$ level. The values calculated from Eq. (5) are, nevertheless, an underestimation of the true pumping efficiency, because the pumping light is absorbed, also, by the atoms in the $F = 2$ sublevel. Then Eq. (4) is modified as follows:

$$A_t = I_c - I_t = K_1\, n_1^t + K_2\, n_2^t, \quad A_p = I_c - I_p = K_1\, n_1^t + K_2\, n_2^t\,, \tag{4'}$$

where the constants $K_1$ and $K_2$ are proportional to the convolution between the pumping light and the absorption line starting from the sublevel $F = 1$ and $F = 2$, respectively. Then, since $n_2^t = \frac{5}{3}\, n_1^t$ and $\Delta n_1 = \Delta n_1$, Eq. (5) is modified as follows:

$$\Delta n_1/n_1^t = [(A_t - A_p)/A_t]\,[(K_1 + \tfrac{5}{3}\, K_2)/(K_1 - K_2)]. \tag{5'}$$

The value of the ratio $K_1/K_2$ is very difficult to calculate but it seems reasonable to put $K_1/K_2 \simeq 3 \sim 5$. Then from Eq. (5′) we find that the true efficiency of the hyperfine pumping is about twice as much as that shown in Fig. 8. Since we can pump the sodium atoms in the upper hyperfine sublevel, it is interesting to investigate the possibility of a Na maser operating between the levels $F = 2$, $m_F = 0 \rightarrow F = 1$, $m_F = 0$.

As compared with the Rb maser there are two advantages: The merit factor $Q$ of the resonant cavity is greater since the frequency of the transition is lower ($Q \propto \sqrt{\lambda}$ ) and the relaxation time is larger since the spin-exchange cross section is smaller.

The threshold condition for maser oscillation is expressed by[17]

$$\alpha N \geq \hbar\, \Delta\nu / 4\mu^2\, Q_0\, \eta \simeq 1.5 \times 10^{12}\, \Delta\nu / Q_0 \eta, \tag{6}$$

where $Q_0$ is the $Q$ of the unloaded cavity, $\eta$ is the filling factor, $\Delta\nu$ is the absorption linewidth, and $\alpha\, N$ is the population difference between the states involved in the transition. Then from Eq. (5′) we find

$$\alpha = \tfrac{1}{8}\, (\Delta n_1/n_1^t)\,(1 + \tfrac{3}{5}) = \tfrac{1}{5}\,(\Delta n_1/n_1^t). \tag{7}$$

A cavity tuned to the Na hyperfine transition has a theoretical $Q_0$ of 72.000 in the TE 011 mode. If the Na vapor completely fills the cavity we can expect a practical $Q_0$ of about $(5–6) \times 10^4$ and a filling factor

$\eta = 0.856,^{18}$ then

$$N \geq 3\times10^{7}\,\Delta\nu/\alpha \simeq 1.4\times10^{10},$$

with $\Delta\nu \simeq 30$ Hz and $\alpha = \frac{1}{15}$ . The result seems quite reasonable, since the pumping efficiency can be further increased either by a better choice of the buffer gas or by increasing the light intensity. Moreover in the resonant cavity of the maser we can expect to have a longer relaxation time, since it must be about three times larger than the cell used in the present investigation.

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[*] Research supported by Gruppo Nazionale di Struttura della Materia del C.N.R.-Sezione di Pisa.

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# Profiles of the He I 4471.5- and 4922-Å Lines in a dc Arc

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Measurements were made of the profiles of the He I 4471.5- and 4922-Å lines in a dc arc for electron densities in the range of 0.4 to $2\times10^{16}$ cm$^{-3}$. Concurrent determinations of the electron densities and temperatures by various methods could not be fully reconciled and are critically discussed. The normalized experimental and theoretical profiles, when compared on the basis of equal peak separation, show increased broadening of the forbidden component and filling in of the dip in the observed profile.

## I. INTRODUCTION

The study of the Stark-broadened spectrum emitted by a plasma has been the subject of considerable research. Recently the He I 4471.5- and 4922-Å line profiles and their forbidden components have been calculated by Griem[1] (4471.5-Å line only) and by Barnard *et al.*[2] and an experiment which provides profiles for critical comparison seemed appropriate. The presented results obtained with a dc arc in an electron density range of 0.4 to $2\times10^{16}$ cm$^{-3}$ should complement those of Nelson and Barnard[3] with a pulsed arc in the range of 3 to $10\times10^{16}$ cm$^{-3}$.

## II. EXPERIMENT

A vortex stabilized helium arc (1.4 atm, 140 A) was viewed side-on using a 3.4-m Ebert mono-

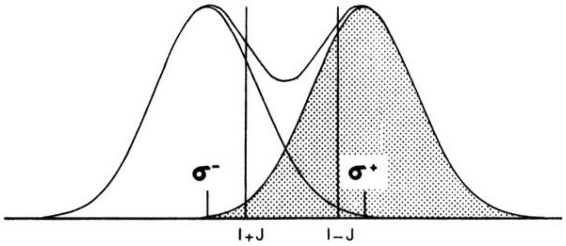

FIG. 1. Zeeman pattern in the field direction. The position of the hyperfine transitions is also shown.

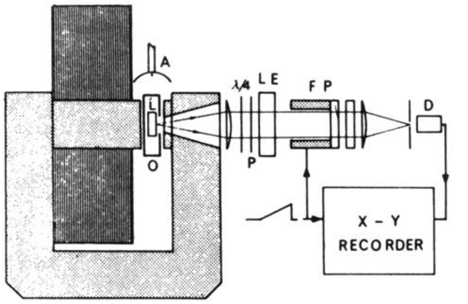

FIG. 2. Experimental apparatus for measuring the shape of the pumping lamp lines. A, microwave antenna; L, Lamp; O, oven; P, polarizer; LE, Lyot-Evans filter; FP, Fabry-Perot interferometer; D, photomultiplier.

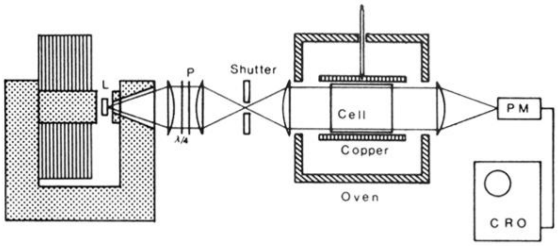

FIG. 4. Experimental apparatus for the hyperfine optical pumping of sodium vapor.