MEASUREMENT OF THE MAGNETIC BIREFRINGENCE OF NOBLE GASES

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ABSTRACT

The Cotton-Mouton constants of argon, krypton, and xenon have been measured, relative to nitrogen, at room temperature and ambient pressure, for $\lambda = 514.5$ nm. Upper limits for helium and neon have been determined.

The sensitivity of the apparatus was $\Delta n_{\min} = 0.8 \times 10^{-14}$ for a measuring time of 1 s.

(Submitted to the Journal of the Optical Society of America)

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1. INTRODUCTION

We present here the first accurate measurements of the magnetic birefringence (Cotton-Mouton effect) in argon, krypton, and xenon. In helium and neon the measured values are within the sensitivity of the apparatus. The measurements have been performed, relative to nitrogen, at room temperature and ambient pressure for two values of the magnetic field intensity and using laser light at $\lambda = 514.5$ nm.

The Cotton-Mouton constant (CM) is given by

$$CM = \Delta n / (\lambda B^2), \quad (1)$$

where B is the component of the magnetic field perpendicular to the light propagation direction, λ is the light wavelength, and $\Delta n = n_{||} - n_{\perp}$ is the induced magnetic birefringence in the medium; $n_{||}$ and $n_{\perp}$ are the refraction indices for light that is linearly polarized parallel and orthogonal, respectively, to the magnetic field direction.

For the gases under study, theoretical estimates [1] give values of the Cotton-Mouton constant which are at least one order of magnitude smaller than those of biatomic molecules such as nitrogen [2-4], for which some of the published values are rather accurate.

The measurements reported here have been carried out with a very sensitive optical ellipsometer, that we are developing also for other purposes [5]. The main features of the apparatus are the following:

- i) The gas birefringence is modulated at a frequency of 1.8 Hz, in order to escape from the high-level noise near zero hertz.
- ii) A heterodyne detection technique is used in which a Faraday modulator acts as a carrier for the birefringence signal.
This method, together with the very high extinction factor obtained, reduces the effect of excess laser noise at low frequency.

In these conditions the sensitivity ψ_{sens} for the measurement of the light ellipticity obtained with the apparatus is

$$\psi_{\text{sens}} = 3.6 \times 10^{-8} / \sqrt{\text{Hz}}. \quad (2)$$

This sensitivity, that is only a factor of 5 worse than the statistical limit at the power level used (60 mW), corresponds to a sensitivity Δn_{min} in the gas birefringence of

$$\Delta n_{\text{min}} = \psi_{\text{sens}} \lambda / (\pi L) = 0.8 \times 10^{-14} / \sqrt{\text{Hz}}, \quad (3)$$

L (80 cm) being the optical path in the birefringent medium.

In the following sections a description is given of the experimental apparatus and of the data-acquisition system. The results of the measurements are given in Section 5. In the Appendix, using a very simple two-level atom model, we show how the Cotton-Mouton constant can be related to the cross-section of the atom-excited state.

2. EXPERIMENTAL APPARATUS

The experimental apparatus is shown in Fig.1. The light source is a CW argon ion laser (Coherent Radiation Mod. CR-2) tuned for $\lambda = 514.5$ nm and for a power output $W \approx 200$ mW. The Glan-Thompson polarizer P (Karl Lambrecht) introduces a well-defined linear polarization state parallel to the horizontal plane. The beam enters a stainless-steel tube T, 4 cm in diameter and 5 m long, which contains the gas under study at 10 mbar above atmospheric pressure. The tube is evacuated to 10^{-3} Torr before each gas filling. The entrance window W to the tube is a BK7 optical flat, 25 mm in diameter, 9 mm thick, mounted with adjustments to minimize the stress birefringence due to pressure differentials. At the opposite end of the tube, the beam is redirected by a spherical mirror M1 ($f = + 5.0$ m) onto the mirror M2. The telescope TL is adjusted so that both the incident and the reflected beam have the waist roughly centred in the entrance window W. The central section of the tube T is placed in the 5 cm gap of the H-type, iron yoke, dipole magnet MD (CERN model NP 300, with modifications). The dipole produces a field of a few kilogauss, sharply confined by field clamps to a region of the tube 40 cm long and uniform to better than 1% in this region. The dipole MD rotates around the beam axis at a frequency of 0.900 Hz, driven by a high-power stepping motor with belt-drive demultiplication. The d.c. excitation current is fed to the dipole coils through slip ring contacts and is monitored via a precision resistor. The pick-up coil PC, placed on the tube T, is used to monitor the modulated field component B orthogonal to the beam propagation direction.

The outgoing light beam from the vessel, reflected by M2, passes through the air Faraday cell FCA2 and is directed to the gold mirror MG. The reflection on the gold mirror transforms the light ellipticity, due to the birefringence of the gas in the modulated magnetic field, into a rotation of the light polarization vector, for a properly chosen incidence angle.

The glass Faraday cell FCG introduces a modulation in the light polarization state which acts as a carrier for the effect to be measured. The cell FCA1 produces a reference rotation of the polarization vector.

The light-beam is directed by the mirror M3 onto the analyser A oriented for maximum extinction. The extinction factor for the crossed prisms P and A is better than 10^{-8} ; however, owing to the presence of the various optical elements and of air, the typical extinction we have obtained with the present apparatus is 10^{-7} , after proper compensation of any static ellipticity through the compensator C.

The transmitted beam is focused on the 2.5 mm diameter PIN photodiode D used in photovoltaic mode and connected to a low-noise current-to-voltage amplifier with feedback resistor of 400 M Ω . This signal is processed by the data-acquisition system described in Section 3. Proper care is taken to reduce any undesired modulated birefringence. For this purpose the tubes containing the gas are well clamped to ground, and the section inside the dipole gap is mechanically isolated by bellows. The entrance window W and the mirror M1 are clamped to the optical bench and are coupled to the tubes by high-resilience bellows. Furthermore, although the residual stray field over the optical elements is not higher than 10^{-4} G, care was taken to place the optical components inside mu-metal shields.

The core of the cell FCG is a 1 cm thick, 2.5 cm diameter BK7 optical flat, placed in a teflon tube at the centre of a 5 cm long solenoid with a

rotation coefficient of 0.19 mrad/A . The power dissipation in the coil for the peak modulation used in the experiment was ≈ 3 W. To ensure dimensional stability and to avoid thermal gradients in the glass core, the coil former was water-cooled. Proper care was taken to avoid transmission of mechanical vibrations from the coil to the glass. The cells FCA1 and FCA2 consist of a solenoid 20 cm long, 2 cm in diameter, wound on a plastic former with a soft-iron sleeve to confine the field return path; their rotation coefficient at room temperature was 4.9×10^{-6} rad/A .

The signal for the Faraday cells together with the trigger for the stepping motor are derived from frequency synthesizers (HP 3325A) locked to the 5 MHz master clock driven by a rubidium maser (HP 5065A). A pulse from the coil SC, derived by inductive coupling from a small permanent magnet fixed to the rotating dipole yoke, is used to synchronize the data acquisition.

The electronic layout is shown in Fig. 2.

3. METHOD AND DATA TAKING

The Cotton-Mouton constants of argon, krypton, and xenon have been measured by comparing their magnetic birefringence with that of nitrogen in the same experimental conditions. According to refs. [3] and [4], we have assumed for nitrogen at 0 °C and 760 Torr the following value of the Cotton-Mouton constant

$$CM(N_2) = -5.10 \times 10^{-17} \text{ G}^{-2} \text{ cm}^{-1}. \quad (4)$$

A linearly polarized light-beam which propagates in a gas where a magnetic field $\underline{B}$ is present will become elliptically polarized, with the major axis of the ellipse rotated with respect to the initial polarization vector. The ellipticity ψ and the rotation ϕ of the ellipse are given by

$$\psi = \pi \text{ CM} \sin(2\theta) \int dx (\underline{B} \wedge \underline{n})^2, \quad (5)$$

$$\phi = V \int dx (\underline{B} \cdot \underline{n}), \quad (6)$$

where the integration is taken along the light path, V is the Verdet constant of the gas, $\underline{n}$ is the unit vector describing the light propagation direction, and θ is the angle between $\underline{B} \wedge \underline{n}$ and the incoming light polarization vector. The intensity transmitted by a crossed analyser prism, to the lowest order in ψ and ϕ , reads

$$I = I_0 (\sigma^2 + \psi^2 + \phi^2), \quad (7)$$

where I_0 is the light intensity prior to the analyser and σ^2 is the extinction factor of the system.

In order to obtain a linear dependence of the signal on ψ , we have introduced an external modulation in the light polarization state. We have preferred a glass Faraday modulator (the cell FCG) to a Pockels cell in order to maintain extinction factors $\approx 10^{-7}$. The choice of the Faraday modulator has required an ellipticity-to-rotation conversion which is obtained by the reflection on the gold mirror MG. We have found that the gold mirror is less noisy in the low-frequency region around 1 Hz than a standard quarter-wave plate.

After the gold mirror, the parameters ϕ' and ψ' which describe the light polarization state are linearly related [6] to the parameters ϕ and ψ before the gold mirror when ϕ and $\psi \ll 1$. One has

$$\phi' = a \psi + b \phi, \tag{8}$$

$$\psi' = a \phi - b \psi,$$

where a and b are parameters which depend only on the complex index of refraction of gold and on the incidence angle. The angles ϕ and ϕ' are measured with respect to the MG incidence plane, which, in our case, is the horizontal plane. The ellipticity-to-rotation conversion would be perfect if $b = 0$. Experimentally we have found

$$a = 1.30 \pm 0.05, \quad |b| < 0.005, \tag{9}$$

for an incidence angle of $\approx 67^\circ$.

Since the two air cells FCA2 and FCA1 produce the same rotation amplitude at two different frequencies, before and after the gold mirror respectively, a measurement of the parameter b is simply given by the ratio between the signals appearing at the two above-mentioned frequencies. During the measurements the attenuation of any Faraday effect prior to the mirror MG was higher than 46 dB.

From eqs.(7) and (8), the light-power on the photodiode is given by

$$\begin{aligned}
 W(t) = W_0 \{ & \sigma^2 + [\phi_g(t) + \phi_{a1}(t) + b \phi_{a2}(t) \\
 & + a \psi_e(t) + b \phi_e(t) + \chi(t)]^2 + \\
 & + [a \phi_e(t) + \rho(t)]^2 \}
 \end{aligned}
 \tag{10}$$

where, in our case,

i) $W_0 \approx 60$ mW is the light-power before the analyser;

ii) $\sigma^2 \approx 1 \times 10^{-7}$ is the over-all extinction factor;

iii) ψ_e is the effect to be measured:

$$\begin{aligned}
 \psi_e &= \Psi_e \sin[2\theta(t)], \quad \theta(t) = 2\pi f_m t, \\
 \Psi_e &= \pi \text{ CM } \int dx B^2, \quad f_m = 0.900 \text{ Hz},
 \end{aligned}$$

(clearly the sign of Ψ_e is positive or negative according to the one of CM);

- iv) ϕ_g is the Faraday rotation angle introduced by the glass Faraday cell FCG:

$$\begin{aligned}\phi_g &= \Phi_g \sin(2\pi f_g t + \zeta_g), \\ \Phi_g &= 3.5 \times 10^{-4} \text{ rad}, \quad f_g = 312.500 \text{ Hz};\end{aligned}$$

- v) ϕ_{a1} is the Faraday rotation angle introduced by the air Faraday cell FCA1:

$$\begin{aligned}\phi_{a1} &= \Phi_{a1} \sin(2\pi f_{a1} t + \zeta_{a1}), \\ \Phi_{a1} &= 0.82 \times 10^{-6} \text{ rad}, \quad f_{a1} = 1.710 \text{ Hz};\end{aligned}$$

- vi) ϕ_{a2} is the Faraday rotation angle introduced by the air Faraday cell FCA2:

$$\begin{aligned}\phi_{a2} &= \Phi_{a2} \sin(2\pi f_{a2} t + \zeta_{a2}), \\ \Phi_{a2} &= 0.82 \times 10^{-6} \text{ rad}, \quad f_{a2} = 1.900 \text{ Hz};\end{aligned}$$

- vii) $\phi_e(t)$ is the Faraday rotation introduced by the residual axial component of the rotating field;

- viii) $\chi(t)$ is the unavoidable optical misalignment ($|\chi| < 10^{-5}$ rad) between the prisms P and A, which may drift slowly in time;

ix) $\rho(t)$ is the residual uncompensated ellipticity ($|\rho| < 10^{-5}$);

with $\zeta_g, \zeta_{a1}, \zeta_{a2}$, initial phases.

The phase of Ψ_e is set to zero by properly positioning the permanent magnet which provides the synchronizing pulse fixing the time origin for data taking.

Since the angles $\Phi_{a1}, \Phi_{a2}, \Psi_e, \Phi_e, \chi$, and ρ are much smaller than Φ_g , the voltage signal $V(t)$ from the current-to-voltage amplifier following D can be written as

$$\begin{aligned}
 V(t) = \eta W(t) R = \eta W_0 R [\sigma^2 + & \frac{\Phi_g^2}{2} - \frac{\Phi_g^2}{2} \cos(4\pi f_g t + 2\zeta_g) \\
 & + 2\Phi_g \chi(t) \sin(2\pi f_g t + \zeta_g) \\
 & + 2\Phi_g \Phi_{a1} \sin(2\pi f_g t + \zeta_g) \sin(2\pi f_{a1} t + \zeta_{a1}) \\
 & + 2a \Phi_g \Psi_e \sin(2\pi f_g t + \zeta_g) \sin(4\pi f_m t) \\
 & + 2b \Phi_g \Phi_{a2} \sin(2\pi f_g t + \zeta_g) \sin(2\pi f_{a2} t + \zeta_{a2}) \\
 & + 2b \Phi_g \Phi_e \sin(2\pi f_g t + \zeta_g) \sin(4\pi f_m t)] ,
 \end{aligned} \tag{11}$$

where η is the photodetector efficiency ($\eta = 0.2$ A/W) and R is the preamplifier feedback resistor (400 M Ω). We have explicitly retained only the second harmonic Fourier component Φ_e of $\phi_e(t)$, since this is the only one which could introduce a systematic error on the measurement of Ψ_e .

In order to evaluate this contribution, we have measured Φ_e in nitrogen with the gold mirror removed, and we found $\Phi_e \approx 10^{-8}$ rad; therefore, taking into account the attenuation given in (9), the contribution of Φ_e becomes smaller than the statistical noise in all our measurements. The signal $V(t)$ is processed by a digital phase-sensitive detector (DPSD) [7] referenced at the modulation frequency f_g , with an output time constant of 100 ms. The reference trigger [7] to the DPSD is adjusted at the start of each measurement to have zero phase difference with respect to the Faraday modulation signal, so that the full demodulated signal power appears in only one output of the DPSD. This procedure is necessary to derive the sign of the Cotton-Mouton constants in the different gases.

Table 1 shows the characteristics of the main Fourier components present in the DPSD output.

For on-line monitoring purposes, we use also a lock-in amplifier (Brookdeal Ortholoc SC 9505E) still referenced at f_g , having an output time constant of 100 ms. The output of the lock-in amplifier and the integrated D signal are processed by a CAVIAR microcomputer. This unit controls the misalignment angle χ and the extinction factor σ^2 by acting, with stepping motors, on the analyser A and the compensator C every 1200 s. The data acquisition is performed by CAMAC units controlled by a HP21MX computer. Each measurement consists of 10 partial runs of 1200 s each, with the field alternately on and off. The first 200 s of each run are allowed for

automatic adjustments of χ and σ^2 . This sequence has been repeated for two values of the magnetic field B , corresponding to 100 A and 150 A of driving current, respectively, in order to verify the quadratic dependence on B of the Cotton-Mouton effect.

The following block of data are recorded on magnetic tape every 50 ms:

- a) DPSP output;
- b) integrated output (0.3 s) from D;
- c) current in the cells FCG, FCA1, and FCA2;
- d) current in the magnet;
- e) signal from the pick-up coil PC.

At the beginning and end of each measurement, we record both the temperature and the pressure of the gas under study.

4. SENSITIVITY OF THE APPARATUS

Several sources of noise and instabilities determine the final sensitivity of the apparatus. The shot-noise in the current of the photodiode D is given, from eq.(10), by

$$\langle i_{\text{shot}} \rangle = \sqrt{2e\eta W_0 (\sigma^2 + \frac{f_g^2}{2}) \Delta\nu} \approx 2.8 \times 10^{-14} \sqrt{\Delta\nu} \text{ A.} \quad (12)$$

The Johnson noise in the 400 M Ω feedback resistor R on the photodiode current-to-voltage preamplifier is

$$\langle i_{\text{thermal}} \rangle = \sqrt{(4kT/R) \Delta\nu} \approx 0.6 \times 10^{-14} \sqrt{\Delta\nu} \text{ A.} \quad (13)$$

The equivalent input noise of the conditioning electronics has been measured to be

$$\langle i_{\text{electr}} \rangle \approx 1. \times 10^{-14} \sqrt{\Delta\nu} \text{ A.} \quad (14)$$

Whilst all these noise sources are "white", the laser intensity fluctuations show a spectral density peaked around zero hertz. Figure 3a shows the Fourier spectrum of the laser intensity between 0 and 10 Hz taken with a low-frequency spectrum analyser (HP 3582A). At 1.8 Hz the laser intensity noise is 25 dB higher than the statistical limit for the same value of d.c. voltage output. The Faraday modulation, acting as a carrier at $f_g = 312.5$ Hz, shifts the signal to a region in which the excess noise of the laser is only 4 dB higher than the statistical limit. Figure 3b shows the laser light intensity spectrum between 0 and 500 Hz. Above 250 Hz, in agreement with refs. [5] and [8], we observe

$$\langle i_{\text{laser}} \rangle \approx 10^{-5} i_{\text{dc}} \sqrt{\Delta\nu}. \quad (15)$$

The good extinction factor of 10^{-7} reached in the apparatus reduces the light-power on the detector to such values that the laser noise becomes comparable with the shot noise; in fact we have

$$\langle i_{\text{laser}} \rangle \approx 2.5 \times 10^{-14} \sqrt{\Delta\nu} \text{ A}. \quad (16)$$

The sum of the noise contributions generates a voltage noise on the signal $V(t)$ from the D preamplifier with a spectral density of

$$V_n \approx 16 \times 10^{-6} \text{ V}/\sqrt{\text{Hz}}. \quad (17)$$

The Faraday modulation Φ_g , according to eq.(11), acts also as a carrier of the fluctuations of the misalignment angle $\chi(t)$, which contribute to the low-frequency noise in the demodulated output (see Fig. 4). These fluctuations are mainly due to stress birefringence induced by thermal gradients in the optical elements and by air convection currents around the beam. To reduce this noise, a protection cover has been fitted to the optical bench.

In order to escape from the near zero hertz noise in the polarization state, the rotating frequency f_m of the dipole has been set to 0.900 Hz, the highest value compatible with its mass (500 kg), the available support, and the driver.

In these conditions, at 1.8 Hz of frequency signal, the sensitivity (2) steadily obtained with the apparatus was five times higher than the statistical limit.

5. DATA ANALYSIS AND EXPERIMENTAL RESULTS

The off-line data analysis is based on the Fourier transform of the recorded DPSD outputs d_n :

$$A_s(f) = \frac{1}{N} \sum_{n=1}^N d_n \sin(2\pi f n \tau), \quad (18)$$

$$A_c(f) = \frac{1}{N} \sum_{n=1}^N d_n \cos(2\pi f n \tau), \quad (19)$$

$$A(f) = \sqrt{A_s^2(f) + A_c^2(f)}, \quad (20)$$

where $\tau = 50$ ms is the sampling time.

Since the start time of the run was determined by the synchronizing pulse from the coil SC, the phase of the effect is zero within 10 mrad. As a consequence, the amplitude $A_c(2f_m)$ does not contain any information about the effect, but only noise. Moreover, $A_s(2f_m)$ and the Cotton-Mouton constant of the gas under study have the same sign. Figure 5 shows a typical Fourier spectrum of the DPSD output, obtained in nitrogen at ≈ 3 kG, for an integration time of 1000 s (frequency resolution 1 mHz). From Table 1 and eq.(18) and (20) we get

$$a \Psi_e / \Phi_{a1} = A_s(2f_m) / A(f_{a1}). \quad (21)$$

Since ϕ_{a1} has a constant peak value of $(8.2 \pm 0.1) \times 10^{-7}$ rad through all the measurements, the determination of $a\psi_e$ is made quite insensitive to the value of the laser intensity, the detector efficiency, and the modulation amplitude ϕ_g .

The experimental results are shown in Table 2, where the two values of the magnetic field are measured by the pick-up coil and correspond to excitation currents of 100 A and 150 A, respectively. The Cotton-Mouton constants for argon, krypton, and xenon are obtained from

$$CM = \frac{A_s(2f_m)}{A(f_{a1})} \frac{A'(f_{a1})}{A'_s(2f_m)} CM(N_2), \quad (22)$$

where A stands for the amplitudes of the gas under study and A' the amplitudes measured in nitrogen in the same conditions.

The statistical error quoted in Table 2 is the r.m.s. value of the Fourier amplitudes A_s near the useful signal frequency (from 1.790 Hz to 1.810 Hz in steps of 1 mHz). The error coming from the uncertainty in ϕ_{a1} is negligible compared to the statistical error.

Analysing the runs performed with field off, we have found a small amplitude at $2f_m$ having a mean value compatible with zero but a r.m.s. value given by

$$\langle A''_s(2f_m) \rangle \approx 0.2 \quad . \quad (23)$$

We have enquired about the origin of these fluctuations, and we think they are due to the residual mechanical vibrations produced by the rotating magnet. We have included the contribution (23) in the total error affecting the Cotton-Mouton constants quoted in Table 2. The results of the measurements performed at 100 A and 150 A are consistent, within the errors.

For helium and neon we have obtained signals comparable to the value (23), which establish the following upper limits for their Cotton-Mouton constants:

$$|\text{CM}(\text{He})| < 6 \times 10^{-19} \text{ G}^{-2} \text{ cm}^{-1}, \quad (24)$$

$$|\text{CM}(\text{Ne})| < 6 \times 10^{-19} \text{ G}^{-2} \text{ cm}^{-1}, \quad (25)$$

with 95% confidence level.

ACKNOWLEDGEMENTS

We thank the technical group of W. Albrecht (Experimental Physics Division, CERN) for making the design of the rotating magnet support, and R. Grabit, F. Jeanmairet, M. Moretti and B. Smith for their technical help.

We are grateful to P. Bernard for the use of laboratory facilities.

APPENDIX

The Cotton-Mouton constant of a monoatomic gas can be evaluated by using a two-level atom model, and related to the linear dimensions of the atom-excited states. The dielectric polarizability tensors [9] χ_{ij} and ρ_{ij} , defined as

$$\underline{D}_i = (\delta_{ij} + 4\pi \chi_{ij}) \underline{E}_j - 4\pi \rho_{ij} \dot{\underline{E}}_j, \quad (A1)$$

can be read as

$$\chi_{ij} = N \frac{2e^2}{\hbar} \sum_s^{\infty} Q_s \sum_{t>s} \frac{\omega_{ts}}{\omega_{ts}^2 - \omega^2} \text{Re } \underline{E}_{ij}^{st}, \quad (A2)$$

$$\rho_{ij} = N \frac{2e^2}{\hbar} \sum_s^{\infty} Q_s \sum_{t>s} \frac{1}{\omega_{ts}^2 - \omega^2} \text{Im } \underline{E}_{ij}^{st}, \quad (A3)$$

where $\underline{E}$, $\underline{D}$, e , $\hbar$, ω_{ts} , and ω have the usual meaning, and

N is the number of atoms per unitary volume;

Q_s is the thermal partition function $Q_s = \exp(-E_s/kT)$, with E_s the energy of the s -state, k the Boltzmann constant, and T the absolute temperature;

ε_{ij}^{st} is defined as

$$\varepsilon_{ij}^{st} = \langle s | \underline{x}_i | t \rangle \langle t | \underline{x}_j | s \rangle, \quad (A4)$$

with $\underline{x}$ the usual position operator.

The tensors ρ_{ij} and χ_{ij} describe the optical activity of the medium and its birefringence, respectively.

Our simplified model of monoatomic gas consists of an S ground state together with a P excited state, separated by an energy difference $\Delta E = \hbar\omega_0$; moreover, only the ground state is populated. Without taking into account the electron spin, in presence of a constant homogeneous magnetic field $\underline{B}$ directed along the z axis, the energy levels are shifted according to Fig. 6. Apart from the Zeeman splitting

$$\omega' = \mu B, \quad \mu = |e|/2mc \quad (A5)$$

of the excited states, the quadratic term in B present in the perturbed Hamiltonian

$$\mathcal{H} = (1/2m) [\underline{p} - (e/2c) \underline{r} \wedge \underline{B}]^2 + V \quad (A6)$$

produces (see Fig. 6)

- a) a shift of the ground state of $\hbar UB^2$;
- b) a shift of $\hbar WB^2$ of the $m = 0$ excited state;
- c) a shift of $2\hbar WB^2$ of the $m = \pm 1$ excited states,

where

$$U = (\alpha/12 mc) \int dr r^4 |R_0(r)|^2 = (\alpha/12mc) \langle r_0^2 \rangle, \quad (A7)$$

$$W = (\alpha/20 mc) \int dr r^4 |R_1(r)|^2 = (\alpha/20mc) \langle r_1^2 \rangle, \quad (A8)$$

α being the fine structure constant, and R_1 and R_0 the radial functions of the excited and ground state, respectively. By assuming this energy-level situation in eq.(A2), it is easy to show that the dielectric polarizability χ_{ij} is such that the only non-zero terms are

$$\chi_{11} = N \alpha c |\langle S | \hat{X} | P \rangle|^2 \left\{ \frac{\omega_1 + \omega'}{(\omega_1 + \omega')^2 - \omega^2} + \frac{\omega_1 - \omega'}{(\omega_1 - \omega')^2 - \omega^2} \right\}, \quad (A9)$$

$$\chi_{22} = \chi_{11}, \quad (A10)$$

$$\chi_{33} = 2 N \alpha c |\langle S | \hat{X} | P \rangle|^2 \frac{\omega_2}{\omega_2^2 - \omega^2}, \quad (A11)$$

where $\langle S | \underline{x} | P \rangle$ is the Wigner-Eckart reduced matrix element [10] of the position operator between the ground state and the excited state, and

$$\omega_1 = \omega_0 + (2W-U)B^2, \quad (A12)$$

$$\omega_2 = \omega_0 + (W-U)B^2. \quad (A13)$$

For propagation of light in the xy plane (i.e. orthogonal to the magnetic field direction), the two indices of refraction $n_{||}$ and $n_{\perp}$ are such that

$$\begin{aligned} n_{||}^2 - 1 &= 4\pi \chi_{33} \cong \\ &\cong (n^2 - 1) \{ 1 - B^2 [(\omega_0^2 + \omega^2)(W-U)] / [\omega_0(\omega_0^2 - \omega^2)] \} , \end{aligned} \quad (A14)$$

$$\begin{aligned} n_{\perp}^2 - 1 &= 4\pi \chi_{11} = 4\pi \chi_{22} \cong \\ &\cong (n^2 - 1) \{ 1 - B^2 [(\omega_0^2 + \omega^2)(2W-U)] / [\omega_0(\omega_0^2 - \omega^2)] \\ &+ \mu^2 B^2 (\omega_0^2 + 3\omega^2) / (\omega_0^2 - \omega^2)^2 \} , \end{aligned} \quad (A15)$$

where n is the index of refraction of the gas, in the absence of external fields. In the same hypothesis, the index n satisfies the equation

$$n^2 - 1 = 8\pi N \alpha c |\langle S | \underline{x} | P \rangle|^2 \omega_0 / (\omega_0^2 - \omega^2) . \quad (A16)$$

According to eqs. (A14) and (A15), the birefringence Δn induced by the magnetic field in the gas is given by

$$\begin{aligned}\Delta n &= n_{||} - n_{\perp} \cong \\ &\cong (n - 1) B^2 \{ [W(\omega_0^2 - \omega^2)] / [\omega_0(\omega_0^2 - \omega^2)] \\ &- \mu^2(\omega_0^2 + \omega^2) / (\omega_0^2 - \omega^2)^2 \} ,\end{aligned}\quad (A17)$$

and therefore, from eq. (1), the Cotton-Mouton constant CM reads

$$CM = \frac{\omega(n-1)}{2\pi c} \left[\frac{W(\omega_0^2 + \omega^2)}{\omega_0(\omega_0^2 - \omega^2)} - \mu^2 \frac{\omega_0^2 + 3\omega^2}{(\omega_0^2 - \omega^2)^2} \right] . \quad (A18)$$

If we put in eq. (A18) the expression of W as given by eq. (A8), and we use for $(n - 1)$ the dispersion relation

$$n - 1 = C / (\omega_0^2 - \omega^2), \quad (A19)$$

we get an equation relating the Cotton-Mouton constant CM with the value of $\sqrt{\langle r_1^2 \rangle}$. Table 3 shows the parameters C and ω_0 which fit the dispersion relation (A19), for noble gases in the visible region (see ref. [11]).

In this way, by using the experimental values of the Cotton-Mouton constants, we have determined the dimension $\sqrt{\langle r_1^2 \rangle}$ of the excited state of the noble gas under study, as obtained in this simplified two-level atom model. In Table 4 we compare the above dimensions with the ones of the corresponding alkali atom in the ground state given, for instance, in ref. [12]. The comparison shows that this very simplified model predicts values which are in rather good agreement with experimental results.

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It turns out that the theoretical results are a factor of 3 smaller than the values that we have found. We think that this discrepancy could be explained by the fact that the authors use the pseudo-potential method to derive the Cotton-Mouton constants of noble gases.

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Table 1: Spectral components of the DPSD output

Spectral component	Frequency (mHz)	Spectral ^{*)} amplitude
d.c.	0	χ
$2f_m$	1800	$a\psi_e$
f_{a1}	1710	$\bar{\phi}_{a1}$
f_{a2}	1900	$b\bar{\phi}_{a2}$

*) The spectral amplitudes are normalized to $2\eta RW_0 \bar{\phi}_g$

Table 2: Experimental results

Gas	B	$A_s(2f_m)$	$A(f_{a1})$	Stat. error	$CM \times 10^{18}$
	(G)	(a.u.)	(a.u.)	(a.u.)	($G^{-2} \text{ cm}^{-1}$)
N ₂	3040	-51.2	184.4	0.13	-51.0 *)
N ₂	4130	-101.3	197.6	0.11	-51.0 *)
Ar	3040	1.26	184.7	0.14	1.25±0.40
					1.32±0.20
Ar	4130	2.39	176.2	0.12	1.35±0.23
Kr	3040	1.77	187.3	0.12	1.74±0.38
					1.93±0.21
Kr	4130	4.00	196.6	0.15	2.02±0.25
Xe	3040	4.43	179.6	0.10	4.53±0.37
					4.45±0.19
Xe	4130	8.98	201.5	0.11	4.43±0.22

*) Value of the Cotton-Mouton constant of nitrogen
assumed according to refs. [3] and [4].

Table 3: Dispersion in noble gases *)

Gas	$C \times 10^{-28}$ (rad/s) ²	$\omega_0^2 \times 10^{-32}$ (rad/s) ²	$(n-1) \times 10^5$ $\lambda = 514.5 \text{ nm}$
He	1.52352	4.39719	3.57
Ne	3.25879	4.89035	6.85
Ar	5.92670	2.13740	29.6
Kr	6.71623	1.60446	45.7
Xe	7.69175	1.12819	77.4

*) Values taken from ref. [11]

Table 4: Atomic radii of noble gases in excited states

Gas	$\text{CM} \times 10^{18}$ $(\text{G}^{-2} \text{ cm}^{-1})$	$\sqrt{\langle r_1^2 \rangle}$ $(\text{\AA})$	Alkali atom	$r_0^{*})$ $(\text{\AA})$
Ar	1.32	2.63	K	2.38
Kr	1.93	2.75	Rb	2.51
Xe	4.45	3.07	Cs	2.70

*) These values are obtained from ref.[12].

FIGURE CAPTIONS

Fig.1 : Experimental apparatus: optical layout.

A: analyser prism; C: compensator;

FCA1, FCA2: air Faraday cells; FCG: glass Faraday modulator;

MG: gold mirror; P: polarizer prism; PC: pick-up coil;

D: photodiode; SC: synchronizing coil; TL: telescope;

W: window; M: manometer; MD: rotating dipole magnet.

Fig.2 : Experimental apparatus: electronic layout.

Fig.3 : Fourier spectrum of the laser intensity fluctuations at

2.5 nA of D current (feedback resistor $R = 400 \text{ M}\Omega$).

The dashed line represents the statistical limit.

The peaks in Fig. 3b correspond to harmonics of the 50 Hz mains.

Fig.4: Fourier spectrum of the lock-in output.

The upper curve is taken with the Faraday modulation on, whereas the lower curve is taken without

Faraday modulation. The magnetic field $\underline{B}$ is off.

The peak at f_{a1} corresponds to the signal produced by the reference cell FCA1 ($\Phi_{a1} = 0.82 \times 10^{-6} \text{ rad}$).

The cut-off near zero hertz is due to the a.c. coupling to the spectrum analyser.

Fig.5 : Fourier amplitude spectrum of the DPSD output, taken in nitrogen at 3040 G of magnetic field intensity, with a measuring time of 1000 s.

Fig.6: Energy-level splittings in the two-level atom model, due to the presence of the magnetic field B.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6