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DOPPLER-FREE TWO-PHOTON SPECTROSCOPY OF NEON
I. FINE STRUCTURE AND HYPERFINE CONSTANTS
FOR THE 4d' SUBCONFIGURATION

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Résumé. — En utilisant la spectroscopie d'absorption à deux photons sans élargissement Doppler, nous avons mesuré la structure fine de la sous-configuration 4d' du néon ainsi que les constantes d'interaction hyperfine des quatre niveaux de cette sous-configuration dans le cas de l'isotope 21. Nous décrivons le montage expérimental et nous insistons sur l'intérêt de placer la cellule d'expérience à l'intérieur d'une cavité Fabry-Pérot accordée à la longueur d'onde excitatrice. Nous comparons nos résultats expérimentaux sur les structures hyperfines avec les prédictions théoriques de Liberman et nous montrons qu'il y a un léger désaccord pour les niveaux de J = 2. Nous expliquons ce désaccord en introduisant une constante d'interaction spin-orbite 64d non nulle pour l'électron externe.

Abstract. — Using Doppler-free two-photon spectroscopy, we have measured the fine structure of the 4d' subconfiguration of neon and the hyperfine constants of the four levels of this subconfiguration for the 21 isotope. We describe the experimental set-up and we point out the interest of putting the experimental cell inside a Fabry-Pérot cavity locked on the laser wavelength. We compare our experimental results for the hyperfine constants with the theoretical predictions of Liberman. We show that there is a slight disagreement for the two J = 2 levels. This is explained by introducing a non zero spin orbit coupling 64d for the external electron.

1. Introduction. — In two previous papers [1, 2], we have presented preliminary results concerning Doppler-free two-photon spectroscopy in Neon. Hyperfine structures in 21Ne and isotope shifts (20Ne, 21Ne, 22Ne) were determined in some of the levels of the 4d configuration.

Since that time other measurements have been performed in this configuration, in cases less favorable than those of the first experiments. Figure 1 gives a simplified energy diagram of Neon showing the concerned levels. These are the four levels of the 4d' subconfiguration labelled 4d' [3/2] 1, 4d' [3/2] 2,

![Simplified diagram of the energy levels of Neon.](image)

(*) Associé au C.N.R.S.

b) The levels of the 4d' sub-configuration.
4d' [5/2] 3, 4d' [5/2] 2 in Racah notation (respectively 4s1, 4s1, 4s1, 4s1 in Paschen notation). An improved experimental set-up with the observed Neon cell inside a Fabry-Pérot cavity coupled with the laser cavity has enabled us to considerably increase the effective irradiating power. Thus we could obtain a fairly good signal-to-noise ratio on levels for which the two photon transition probability would be too small for observation with the less sophisticated set-up.

As the isotope shift results for these levels have already been presented and discussed in ref. [2], we are going to deal here with fine and hyperfine structures; after describing the experimental set-up, we will give the results of our measurements and compare them to the values derived by a semi-empirical method [3]. We will show how comparison between theory and experiment lead us to propose an improvement to the theory.

2. Experimental set-up. — The main features of a Doppler-free two-photon experiment are now well-known: the sample — here the neon cell — is irradiated by a monomode laser beam which is reflected back onto itself, so that an atom moving with axial velocity $v$ sees two waves with frequencies $\omega(1 - v/c)$ and $\omega(1 + v/c)$ ($\omega$ being the laser frequency). Thus the condition for the atom to be excited from level a to level b by absorbing two photons (one from each wave) is, at resonance

$$\omega_b - \omega_a = \omega(1 - v/c) + \omega(1 + v/c) = 2 \omega$$

which is independent of atomic velocity: the Doppler effect is eliminated.

Our experimental set-up (Fig. 2) involves a c.w. laser, the frequency of which is scanned by the means of servo-systems. The Spectra-Physics (model 580) dye laser pumped by a 4WAr+ laser (5 145 Å) can deliver about 150 mW for the resonance wavelengths (around 5 920 Å) in monomode operation, which is enough for our experiments. Monomode running is provided by an internal Fabry-Pérot etalon, the thickness of which can be piezoelectrically varied.

One of the laser mirrors is also mounted on a piezoelectric transducer. Two servo-loops are used. The first one locks the frequency determined by the internal etalon on one mode of the laser cavity, using a modulation of the thickness of this etalon. The second one locks the length of the laser cavity on the side of the transmission peak of an external planar Fabry-Pérot etalon (free spectral range: 5 GHz); this loop involves comparison between the overall laser intensity and the intensity transmitted through the etalon, and does not necessitate any modulation. Due to the remaining jitter, the laser mode has a width of about 7 MHz in a time of the order of a fraction of a second. The external etalon is thermally very stable, because of its ceramic spacers; it is located inside a vacuum box; letting air in through a calibrated leak provides pressure scanning of the laser frequency. Such a frequency scanning is almost linear; moreover the non-linear corrections can be easily calculated if necessary.

To calibrate the frequency scanning, a planar-focal etalon with a free spectral range of 75 MHz is used. It is constructed with Invar and thermostated to 0.01 °C.

These stabilizing and monitoring devices only take a small part of the intensity. The main beam is focussed inside the experience cell using a 15 cm lens and reflected back on itself by a spherical mirror. An optical isolator is placed on the way of the laser beam to avoid coupling between the return beam and the laser cavity. We have used either a Faraday rotator, a glass rod (Sovirel, flint E0525) submitted to a convenient magnetic field when working with linear polarization, or a quarter-wave plate when circular polarization was needed. A further improvement consisted in making the laser beam pass several times through the cell: the cell is then placed inside a concentric resonator formed with two spherical mirrors; the rear mirror has a 99.5 % reflectance, the input mirror has a 94 % reflectance. A 20 cm lens is used to match this cavity to the laser cavity. One of the mirrors is mounted on a piezoelectric ceramic. The length of the cavity is modulated and a servo-loop is used to lock the cavity length so that the transmitted light is maximum. The interest of performing the experiment in such a cavity is clearly demonstrated in figure 3, where we show recordings of the $3s \ J = 2 \rightarrow 4d' \ J = 1$ two-photon resonance without (Fig. 3a) and with the Fabry-Pérot resonator (Fig. 3b). The signal to noise ratio is increased by a factor of 15.

The neon cell (1) has been sealed off after filling. As can be seen in figure 1a the two-photon transitions concerned in the experiment do not start from the ground state but from a metastable state, that we must populate with a discharge. But the discharge brings atoms in all the other excited states too, making

(1) To minimize the reflection losses we use a cell with Brewster-angle windows; inside the Fabry-Pérot resonator we can only work with linear laser polarization.
it impossible to detect the two-photon signal out of the fluorescence noise; so we had to work with an after-glow, where only the metastable states remain populated. A chopped discharge is supplied by a 14 MHz generator, the output of which is modulated in square wave at a frequency of a few kHz (the lifetime of the metastable state is of the order of one millisecond in our cells). The fluorescence light emitted on one of the transitions from the excited state under investigation to a lower state is selected with a monochromator and detected with a photomultiplier. To eliminate the fluorescence signal of the discharge when it is on, the photomultiplier current is only transmitted during the after-glow, using a SHA 1A (Analog Devices) amplifier. When sweeping the laser frequency the resulting signal is recorded as a function of time with a 3 s time-constant on the first trace of a double trace recorder. The transmission peaks given by a 75 MHz reference etalon are simultaneously recorded on the second trace. Such a recording is shown in figure 4.

The last column of table I gives the values derived from Ch. Moore’s tables. It can be seen that the accuracy is considerably improved. Moreover, if the accuracy of the values from the spectroscopic tables is considered to be 1.0 mK, our values are not in agreement with them.

Because of the order of magnitude of these frequency intervals, it was not possible to cover them in a continuous scan of the laser frequency (the piezo-electric ceramic carrying the laser mirror only permits amplitudes of a few GHz). The experiments has been done by sweeping the laser frequency over a small range around each two-photon resonance and by changing it by steps and counting the orders of convenient Fabry-Pérot etalons between.

3. Experimental results. — 3.1 Fine structure intervals. — The energy separations between the four levels of the 4d’ subconfiguration are reported in table I. Let us point out that these results have been obtained on 20Ne isotope with a natural neon pressure of 0.6 torr. Because of the specific mass-shift, the results would be different for 22Ne [2]. On the other hand, one must take into account the pressure shift of the two-photon line due to interatomic collisions. The detailed investigation of these pressure shifts [5] has shown that they are nearly the same for the four 4d’ levels; so we do not need to extrapolate the fine structure results to zero pressure.

Table I

<table>
<thead>
<tr>
<th>This work</th>
<th>Previous values [9]</th>
</tr>
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<tbody>
<tr>
<td>4d'[5/2] 3-4d'[5/2] 2</td>
<td>924.9 ± 0.2 mK</td>
</tr>
<tr>
<td>4d'[3/2] 2-4d'[5/2] 3</td>
<td>1 051.6 ± 0.2 mK</td>
</tr>
<tr>
<td>4d'[3/2] 1-4d'[5/2] 3</td>
<td>11 869.2 ± 1 mK</td>
</tr>
</tbody>
</table>

The 3.2 Hyperfine structure intervals. — Using a cell containing 99 % enriched 21Ne isotope, we have recorded the various hyperfine components of the two-photon transitions. A typical recording is shown in figure 4. The hyperfine structure of the metastable 3s[3/2] 2 (1s5) level is well-known [6] and we have used it to verify the frequency calibration given by the Fabry-Pérot etalon. Nearly all the components are well separated. It was rather easy to identify them because, as shown in ref. [7], it is possible to predict their relative intensities, and because we already knew the order of magnitude of the hyperfine structures from ref. [3]. Table II shows the hyperfine intervals for the four investigated levels.

4. Interpretation of the hyperfine structures. — 4.1 Calculation of the dipolar magnetic and quadrupolar electric constants A(αJ) and B(αJ). — As the hyperfine interaction is not very small
TABLE II
Hyperfine structure intervals in the 4d' subconfiguration

| 4d' [5/2] 2 | 5/2-7/2 | 1 233 ± 4 MHz |
| 3/2-5/2 | 902.5 ± 10 MHz |
| 1/2-3/2 | 534.0 ± 10 MHz |
| 7/2-9/2 | 998.8 ± 2.6 MHz |
| 5/2-9/2 | 1 832.2 ± 3.0 MHz |
| 3/2-9/2 | 2 452.2 ± 3.7 MHz |
| 4d' [3/2] 2 | 7/2-5/2 | 818.8 ± 6.6 MHz |
| 5/2-3/2 | 602.6 ± 7.7 MHz |
| 3/2-1/2 | 375.4 ± 8.2 MHz |
| 4d' [3/2] 1 | 5/2-3/2 | 873.8 ± 3.4 MHz |
| 3/2-1/2 | 522.8 ± 3.3 MHz |

compared to the fine structure intervals in the 4d' sub-configuration, the hyperfine coupling between levels 4d' [5/2] 3, 4d' [5/2] 2, 4d' [3/2] 2 and 4d' [3/2] 1 cannot be neglected. So the various hyperfine sublevels are shifted from the position they would have if one only took into account the diagonal part of the hyperfine interaction. To correctly interpret the experimental hyperfine intervals, we have to introduce a corrective term in Casimir formula, and write the energy of sublevel $F$ of level $aJ$ in the following form:

$$E(aJF) = A(aJ) \left( \frac{C_F}{2} + B(aJ) \right) + \delta E(aJF)$$

where

$$C_F = F(F + 1) - (I + 1) - J(J + 1)$$

with $I = 3/2$ for $^{21}$Ne.

The expression of $\delta E(aJF)$ can be determined from second order perturbation theory with respect to the non-diagonal part of the hyperfine hamiltonian $\mathcal{H}_{\text{hyp}}$.

$$\delta E(aJF) = \sum_{aJ'} | \langle aJFM | \mathcal{H}_{\text{hyp}} | a'J'F'm \rangle |^2.$$  

The summation being done over the three other states of the 4d' subconfiguration.

Using the tables of eigenstates of Neon given in [4], we see that the studied 4d' states are not very different from pure states of Racah's basis (2).

The components of the eigenstate labelled $| 4d' [K] J \rangle$ on states of Racah's basis with $j_1 \neq 1/2$ or $K' \neq K$, as given in [4], are small, and it can be shown that their contribution to $\delta E(aJF)$ is negligible in comparison with our experimental precision; so for this calculation of $\delta E(aJF)$, we shall only take the main component into account.

As the external electron is in a rather excited level with non-zero momentum, the hyperfine hamiltonian is only due to the interaction between the nucleus and the core electrons. Moreover, the quadrupolar hyperfine interaction has no matrix elements between states with $j_1 = 1/2$ so we only have to compute the matrix elements of the dipolar hamiltonian

$$\mathcal{H}_D = X I.$$  

$X$ is an electronic vectorial operator, so $\mathcal{H}_D$ is diagonal with respect to $F$ and $m_F$.

The matrix elements involved in the calculation of $\delta E(4d' [K] JF)$ can finally be written as:

$$\langle 4d' KJFM_F | X I | 4d' K' J' F'M_{F'} \rangle = c_1(K, J, K', J', F) \langle j_1 \| X \| j_1 \rangle$$

with $j_1 = 1/2$.

The expression for $c_1$ is derived in the appendix. On the other hand, the magnetic dipolar hyperfine constant $A(4d' [K] J)$ can also be expressed as a function of $\langle j_1 \| X \| j_1 \rangle$.

$$A(4d' [K] J) = = c_2(K, J) \langle j_1 \| X \| j_1 \rangle$$

(see appendix for the exact expression of coefficient $c_2$).

Using the value of $A(4d' [5/2])$ given by the theory of ref. [3], we obtain the value of $\langle j_1 \| X \| j_1 \rangle$ for $j_1 = 1/2$ and we can then calculate the matrix elements involved in $\delta E(4d' [K] JF)$. The $\delta E$ values are quite important; for example in level 4d' [5/2] 3, the $F = 3/2, F = 5/2$ and $F = 7/2$ are respectively shifted of $- 27$ MHz, $- 54$ MHz and $- 54$ MHz. From the so corrected hyperfine intervals, the Casimir formula enables the $A$ and $B$ values to be determined as given in table III. The last column shows the values derived from theory. As far as the magnetic structures are concerned, the agreement is fairly good for level 4d' [5/2] 3 (which justifies the value of $A(4d' [5/2] 3)$ chosen for the calculation of $\langle j_1 \| X \| j_1 \rangle$) and for level 4d' [3/2] 1; for the $J = 2$ levels the agreement is not so good and we propose an interpretation of this fact in the next paragraph. For the quadrupolar part, in view of the error bars, no significant feature can be deduced from the agreement between theory and experiment.

4.2 INTERPRETATION OF THE DISAGREEMENT BETWEEN THEORY AND EXPERIMENT FOR THE $J = 2$ LEVELS.

As pointed out before, the wave functions derived...
Table III

Hyperfine structure constants

<table>
<thead>
<tr>
<th></th>
<th>(A_{\text{exp}}) (MHz)</th>
<th>(B_{\text{exp}}) (MHz)</th>
<th>(A_{\text{c}})</th>
<th>(B_{\text{c}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>4d' [5/2] 2</td>
<td>-355.4 ± 1.2</td>
<td>+8 ± 5</td>
<td>-327.3</td>
<td>+8.1</td>
</tr>
<tr>
<td>4d' [5/2] 3</td>
<td>{-235.9 ± 0.5 }</td>
<td>+11 ± 9</td>
<td>-234.9</td>
<td>+6.6</td>
</tr>
<tr>
<td>4d' [3/2] 2</td>
<td>+231.4 ± 1.6</td>
<td>+2 ± 6</td>
<td>+206.1</td>
<td>+5.7</td>
</tr>
<tr>
<td>4d' [3/2] 1</td>
<td>+348.3 ± 1.6</td>
<td>+0.8 ± 1.7</td>
<td>+345.0</td>
<td>+2.4</td>
</tr>
</tbody>
</table>

(1) These two values correspond to different ways of calculating the second order corrections; for the \(A\) and \(B\) values of the first line, the second order corrections have been calculated following § 4.1 with the eigenstates of ref. [4]; for the second line the second order corrections have been calculated by taking into account § 4.2.

We have neglected the cross terms coming from the small components of states with \(j_z = 3/2\). A similar expression can be written for \(A(4d'[3/2] 2)\).

Using the hyperfine interaction parameters of ref. [3] and replacing \(A(4d'[5/2] 2)\) and \(A(4d'[3/2] 2)\) by their experimental values, we obtain two determinations of \(\theta\) which agree with each other: \(\theta_1 = 11.6^\circ\) and \(\theta_2 = 11.7^\circ\); hence the mixing coefficients are obtained

\[
\sin \theta = 0.20, \quad \cos \theta = 0.98.
\]

It can easily be shown that the non-diagonal element of the spin-orbit interaction between the states \(|1/2, 5/2, 2\rangle\) and \(|1/2, 3/2, 2\rangle\) is \(\sqrt{6}/5 \xi_{4d}\). Using our experimental value of the energy difference between levels 4d' [5/2] 2 and 4d' [3/2] 2 we deduce

\[
\xi_{4d} = 0.8 \text{ cm}^{-1}.
\]

This value is small in comparison with the parameters involved in the electrostatic interaction and the spin-orbit coupling of the core. It only leads to important effects in the 4d configuration because of the small energy differences between the levels. Let us note that this interpretation of the mixing of the wave-functions by the spin-orbit coupling of the external electron is only an approximation. It has been shown that other interactions (spin-spin and spin-other orbit for example) can play a role in the 4d configuration [8]. Nevertheless, these interactions are always weaker than that due to spin-orbit and the order of magnitude we obtain for \(\xi_{4d}\) remains valid.

Remark. — We have just shown that the wave-functions given in ref. [4] have to be modified by non-negligible quantities. One can then think that the corrections \(\delta E(4d'[K] J)\) to the hyperfine structure intervals calculated in the preceding paragraph are no longer valid and thus that the experimental hyperfine structure constants must be recalculated. In fact, one finds modifications which are small compared to the error bars except for the 4d' [5/2] 3 level for which the result is now:

\[
A = -236.3 \pm 0.5, \quad B = 6 \pm 11.
\]

5. Conclusion. — We have measured the hyperfine constants of the four levels of the 4d' subconfiguration. These measurements have provided new information concerning the excited states wave-functions and on the spin-orbit coupling in these excited states. Furthermore, the intensities of the hyperfine components provide a very accurate test of the theory of two-photon excitation. The corresponding results are presented in a following paper.
Appendix. — As we have neglected the hyperfine interaction between the external electron and the nucleus, $X$ is a vectorial operator which only acts on the core electrons; using the properties of the $6J$ coefficients we have:

$$
\langle 4d' \, KJ\!\!J'\!\!J' \mid X \mid 4d' \, K'\!\!J' \!\!J' \rangle = \sqrt{15 \left( (-)^{F+1/2+K+K'} \right) \sqrt{2 K + 1} \sqrt{2 K' + 1} \sqrt{2 J + 1} \sqrt{2 J' + 1} \times \\
\times \begin{cases}
1/2 & 1 \\
K & 1 \\
K' & 2 \\
K & \end{cases} \begin{cases}
J & 3/2 \\
J' & 3/2 \\
F & \end{cases} \left\langle j_1 = 1/2 \parallel X \parallel j_1 = 1/2 \right\rangle
$$

where we have taken $I = 3/2$.

Similarly, the usual coupling formulas enable us to write the magnetic dipolar hyperfine constant as a function of the reduced matrix element $\left\langle j_1 \parallel X \parallel j_1 \right\rangle$

$$
A(4d' [K] J) = \left[ (J + 1) (2 J + 1) \right]^{-1/2} \langle 4d' \, KJ \parallel X \parallel 4d' \, KJ \rangle \approx
$$

$$
\approx \left[ (J + 1) (2 J + 1) \right]^{-1/2} (-)^{F} (2 K + 1) (2 J + 1) \begin{cases}
1/2 & 1 \\
K & 1 \\
K & 2 \\
K & \end{cases} \begin{cases}
1/2 & 1 \\
J & 1/2 \\
J & \end{cases} \times
$$

$$
\times \left\langle j_1 = 1/2 \parallel X \parallel j_1 = 1/2 \right\rangle
$$

which gives the coefficients $c_1$ and $c_2$.

References