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Doppler-free two-photon spectroscopy of neon. III. Isotopic shift and fine structure for the $2p^5 4d$ and $2p^5 5s$ configurations

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Résumé. — En utilisant la spectroscopie d'absorption à deux photons sans élargissement Doppler, nous avons mesuré l'écart isotopique ^{20}Ne - ^{22}Ne pour des transitions reliant le niveau métastable $3s[3/2]2$ et les niveaux excités des configurations $2p^5 4d$ et $2p^5 5s$. Les résultats sont analysés en utilisant la théorie du déplacement de masse spécifique. Nous montrons que nous pouvons déduire des données expérimentales un développement du terme 1P de Russel-Saunders sur les états propres du hamiltonien atomique. Nous obtenons également des déterminations expérimentales des intégrales radiales $J(4d, 2p)$ et $J(5s, 2p)$. Les résultats expérimentaux sont en bon accord avec des prévisions théoriques de Bauche et Liberman.

Abstract. — Using Doppler-free two-photon spectroscopy, we have measured the isotopic shift ^{20}Ne - ^{22}Ne for two-photon transitions between the $3s[3/2]2$ metastable state and the excited states of the $2p^5 4d$ and $2p^5 5s$ configurations. The results are analyzed using the specific mass shift theory. We show that we can deduce from the experimental data the expansion of the 1P Russel-Saunders term over the true eigenstates of the atom. We also obtain experimental determinations of the radial integrals $J(4d, 2p)$ and $J(5s, 2p)$. All the experimental results are in good agreement with theoretical predictions of Bauche and Liberman.

Doppler-free two-photon spectroscopy has provided considerable information about many atoms and molecules [1]. The experiments performed on neon [2, 7] are perhaps the most evident demonstration of the power of the method. Using a c.w. dye laser of moderate power, it has been possible to measure many fine and hyperfine structures [2, 4, 6], to determine isotopic shifts [2, 3] and to obtain much valuable information about two-photon line intensities [5]. In the present paper, the Doppler-free two-photon excitation method is applied to measure the isotopic shifts ^{20}Ne - ^{22}Ne and the fine structure in the $2p^5 4d$ and $2p^5 5s$ configurations of neon. Some preliminary results about the isotopic shift have already been published [3], but the measured values were restricted to the levels of the $4d'$ subconfiguration. The measurement of all the isotopic shifts of the $2p^5 4d$ and $2p^5 5s$ configurations brings new information and now permits a precise comparison

with theory. It has been possible to obtain valuable information on the wave functions of the levels of angular momentum $J = 1$: the $J = 1$ levels have a specific mass-shift different from that of the other levels, and this shift depends on their wavefunctions. The experiments also provide a determination of the radial integrals $J(4d, 2p)$ and $J(5s, 2p)$. Our measurements in the $4d$ and $5s$ configurations give wavefunctions in agreement with predictions of Liberman [8] and radial integrals in agreement with predictions of Bauche [9]. The paper is divided in three parts. In the first part we describe the problems involved in the excitation of the levels of the $4d$ and $5s$ configurations. In part 2 we give the experimental results for the isotopic shifts and for the fine structures. In the last part we make a theoretical analysis of the experimental data and we show the quantities which can be deduced from them.

1. **Some preliminary remarks about the two-photon transitions presented in this paper.** — We will not describe in this paper the experimental set-up. It is identical to that described in ref. [4]; in particular all the experiments have been performed with a neon cell placed inside a concentric resonator.

We have observed the two-photon transitions between the $3s[3/2]2$ (Racah notations) metastable state of neon and all the levels of the $2p^5 4d$ and $2p^5 5s$ configurations. The metastable state is populated using a discharge. The laser wavelengths for the two-photon excitation are given in table I. The levels of the $2p^5 4d$ configuration were excited using a Rhodamine 6G dye laser pumped by the 514 nm line of an argon ion laser while the levels of the $2p^5 5s$ configuration were excited using a Rhodamine 640 dye laser pumped by the green-yellow lines of a krypton ion laser. We have also reported in table I the fluorescent lines we used to detect the various resonances.

Table I.

Two-photon transition	λ_{ex} (Å)	Detection	λ_{det} (Å)
$3s[3/2]2-4d'[3/2]1$	5 921.5	$4d'[3/2]1-3p'[1/2]1$	5 961
$3s[3/2]2-4d'[3/2]2$	5 923.4	$4d'[3/2]2-3p'[1/2]1$	5 965
$3s[3/2]2-4d'[5/2]2$	5 923.8	$4d'[5/2]2-3p'[3/2]1$	5 872
$3s[3/2]2-4d'[5/2]3$	5 923.6	$4d'[5/2]3-3p'[3/2]2$	5 902
$3s[3/2]2-4d[1/2]0$	6 072.58	$4d[1/2]0-3p[3/2]1$	5 934
$3s[3/2]2-4d[1/2]1$	6 071.16	$4d[1/2]1-3p[1/2]1$	5 341
$3s[3/2]2-4d[3/2]1$	6 061.66	$4d[3/2]1-3p[3/2]1$	5 913
$3s[3/2]2-4d[3/2]2$	6 064.50	$4d[3/2]2-3p[1/2]1$	5 330
$3s[3/2]2-4d[5/2]2$	6 057.87	$4d[5/2]2-3p[5/2]2$	5 804
$3s[3/2]2-4d[5/2]3$	6 057.68	$4d[5/2]3-3p[5/2]3$	5 748
$3s[3/2]2-4d[7/2]3$	6 066.42	$4d[7/2]3-3p[5/2]2$	5 820
$3s[3/2]2-4d[7/2]4$	6 066.61	$4d[7/2]4-3p[5/2]3$	5 764
$3s[3/2]2-5s'[1/2]0$	6 139.96	$5s'[1/2]0-3p[3/2]1$	6 113
$3s[3/2]2-5s'[1/2]1$	6 130.51	$5s'[1/2]1-3p[3/2]2$	6 328
$3s[3/2]2-5s[3/2]1$	6 273.57	$5s[3/2]1-3p[5/2]2$	6 214
$3s[3/2]2-5s[3/2]2$	6 290.27	$5s[3/2]2-3p[1/2]1$	5 690

Table II. — *Relative transition probabilities (arbitrary units).*

Two-photon transition	$ \langle e \parallel q^0 \parallel g \rangle ^2$	$ \langle e \parallel q^2 \parallel g \rangle ^2$	$\frac{1}{3} \langle e \parallel q^2 \parallel g \rangle ^2 + \frac{2}{3} \langle e \parallel q^2 \parallel g \rangle ^2$
$3s[3/2]2-4d'[3/2]1$	0	22×10^{-8}	14.6×10^{-8}
$3s[3/2]2-4d'[3/2]2$	0.1×10^{-8}	80×10^{-8}	53×10^{-8}
$3s[3/2]2-4d'[5/2]2$	14×10^{-8}	28×10^{-8}	23.4×10^{-8}
$3s[3/2]2-4d'[5/2]3$	0	218×10^{-8}	145×10^{-8}
$3s[3/2]2-4d[1/2]0$	0	2×10^{-11}	1.3×10^{-11}
$3s[3/2]2-4d[1/2]1$	0	0.69×10^{-8}	0.45×10^{-8}
$3s[3/2]2-4d[3/2]1$	0	0.44×10^{-8}	0.30×10^{-8}
$3s[3/2]2-4d[3/2]2$	29.4×10^{-8}	13.5×10^{-8}	18.8×10^{-8}
$3s[3/2]2-4d[5/2]2$	8×10^{-11}	0.6×10^{-8}	0.4×10^{-8}
$3s[3/2]2-4d[5/2]3$	0	38×10^{-8}	25×10^{-8}
$3s[3/2]2-4d[7/2]3$	0	1.8×10^{-8}	1.2×10^{-8}
$3s[3/2]2-4d[7/2]4$	0	16×10^{-8}	10.6×10^{-8}
$3s[3/2]2-5s'[1/2]0$	0	16×10^{-8}	10.6×10^{-8}
$3s[3/2]2-5s'[1/2]1$	0	175×10^{-8}	116×10^{-8}
$3s[3/2]2-5s[3/2]1$	0	88.9×10^{-8}	59.3×10^{-8}
$3s[3/2]2-5s[3/2]2$	46.6×10^{-8}	5.9×10^{-8}	19.5×10^{-8}

The transition probabilities are very different from one transition to another. These transition probabilities can be calculated using the results of ref. [5]. From formulae (17) and (34) of ref. [5], it can be shown that with a linear polarization of the exciting light, the transition probability P_{ge} is proportional to :

$$\frac{1}{3} |\langle e \parallel q^0 \parallel g \rangle|^2 + \frac{2}{3} |\langle e \parallel q^2 \parallel g \rangle|^2$$

where q^0 and q^2 are the scalar and quadrupolar parts of the two-photon transition operator (which involves a bilinear product of the electric dipole operator). With circular laser polarization only the quadrupolar part of the two-photon operator is involved and the transition probability is proportional to

$$|\langle e \parallel q^2 \parallel g \rangle|^2.$$

The values of the reduced matrix elements of the scalar and quadrupolar operators q^0 and q^2 can be calculated using formula (35) of ref. [5]. The corresponding values are reported in table II. From this table it can be seen that the transition probabilities vary by five orders of magnitude from the most intense two-photon transition $3s[3/2]2-4d'[5/2]3$ (which was observed first in [2]) to the less intense one $3s[3/2]2-4d[1/2]0$. Nevertheless even this very small two-photon resonance has been experimentally observed. It can also be noticed that the relative intensities of the experimental signal are sometimes different from what can be expected from the two-photon transition probability. The main reason for it is that it is not always possible to detect the resonance on the most intense fluorescent line (because it lies in the vacuum U.V. range or because it is too close to the laser wavelength for the monochromator to cut off all the laser stray light). Another reason is that we have assumed in our calculations that the ground and excited states are Racah states ; but it can be

shown that some of these states (for instance $4d'[3/2]2$ and $4d'[5/2]2$) slightly differ from pure states in Racah's coupling.

2. Experimental results. — All the experiments have been performed with a cell containing 0.6 torr of natural neon. Each transition consists of two lines distant by about 2.6 GHz, corresponding to the isotopes 20 Ne and 22 Ne (see Fig. 1). The line cor-

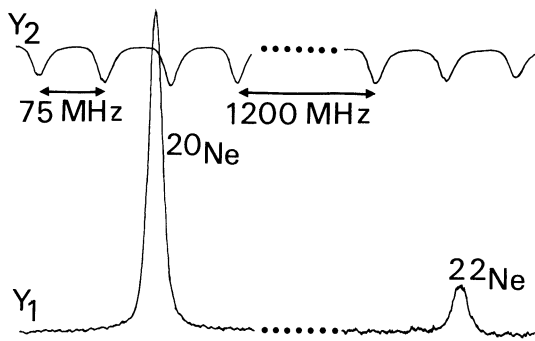


Fig. 1. — Experimental recording of a Doppler-free two-photon transition in a pure neon cell (case of the $3s[3/2]2-4d'[5/2]3$ transition). The linewidth of the transition is partly due to collisional broadening (the recording has been performed with a neon pressure of 0.6 torr). The upper trace corresponds to the simultaneous recording of a Fabry-Perot etalon of free-spectral range equal to 75 MHz which corresponds to 150 MHz on the transition frequency.

responding to 20 Ne is about ten times larger than the line corresponding to 22 Ne, due to natural isotopic abundance. From the relative positions of the two-photon transitions reaching the $2p^5 4d$ and $2p^5 5s$ levels, it has been possible to deduce the fine struc-

Table III. — *Fine structure intervals in the 4d and 5s configurations.*

Interval	Measured value (cm ⁻¹)	Ch. Moore's table
$4d'[3/2]1-4d'[5/2]3$	11.869 2(10)	11.865
$4d'[3/2]2-4d'[5/2]3$	1.051 6(2)	1.050
$4d'[5/2]3-4d'[5/2]2$	0.924 9(2)	0.931
$4d[1/2]1-4d[1/2]0$	7.667 1(4)	7.682
$4d[7/2]4-4d[1/2]0$	32.356 4(4)	32.368
$4d[7/2]3-4d[1/2]0$	33.457 0(3)	33.465
$4d[3/2]2-4d[1/2]0$	43.889 0(3)	43.896
$4d[5/2]1-4d[1/2]0$	59.317 2(10)	59.318
$4d[5/2]2-4d[1/2]0$	79.933 1(3)	79.941
$4d[5/2]3-4d[1/2]0$	80.994 5(3)	81.000
$5s[3/2]1-5s[3/2]2$	84.609 8(3)	84.612
$5s'[1/2]1-5s'[1/2]0$	50.174 4(3)	50.175

tures in these two configurations. The measurements reported in table III correspond to 20 Ne and to a pressure of 0.6 torr. The results would be different for 22 Ne because of the specific mass-shift. We did not perform a systematic study of the pressure shift; nevertheless, from the results about the pressure shift obtained in the $4d'$ subconfiguration [10], and from what is theoretically expected in the other levels, we believe that at 0.6 torr the difference in the values of the pressure shift is smaller than our error bar. Our values in table III are compared with the values derived from Ch. Moore's table [11]. It can be seen that the accuracy is considerably improved.

As can be seen in figure 1, the experimental recording provides the isotopic shift of a Doppler-free two-photon transition very directly. The values of the isotopic shifts on the metastable-excited state two-photon transitions are reported in tables IV and V

Table IV. — *Analysis of the experimental results for the $2p^5 4d$ configuration (all the data are in MHz).*

Two-photon transition	Isotopic shift $^{20}\text{Ne}-^{22}\text{Ne}$ experimental value Δ_{exp}	Bohr mass shift (calculated) Δ_B	Specific shift $\Delta_S = \Delta_{\text{exp}} - \Delta_B$	Shift of $J=1$ levels $\Delta_S(J=1) - \Delta_S(J \neq 1)$	Specific shift due to the subconfiguration $\overline{\Delta_S}(j_1=1/2) - \overline{\Delta_S}(j_1=3/2)$
$3s[3/2]2-4d'[3/2]1$	$2\ 764.6 \pm 3.0$	2 526.1	238.5 ± 3.0	$- 16.6 \pm 4.4$	} 23.2 ± 2.8
$3s[3/2]2-4d'[3/2]2$	$2\ 779.2 \pm 4.0$	2 525.3	253.9 ± 4.0		
$3s[3/2]2-4d'[5/2]2$	$2\ 781.8 \pm 2.0$	2 525.2	256.6 ± 2.0		
$3s[3/2]2-4d'[5/2]3$	$2\ 780.2 \pm 1.6$	2 525.3	254.9 ± 1.6		
$3s[3/2]2-4d[1/2]1$	$2\ 686.0 \pm 2.6$	2 463.9	222.1 ± 2.6	$- 9.8 \pm 4.0$	}
$3s[3/2]2-4d[3/2]1$	$2\ 668.7 \pm 5.0$	2 467.7	201.0 ± 5.0	$- 30.9 \pm 6.4$	
$3s[3/2]2-4d[3/2]2$	$2\ 699.3 \pm 2.1$	2 466.6	232.7 ± 2.1		
$3s[3/2]2-4d[5/2]2$	$2\ 702.5 \pm 5.0$	2 469.3	233.2 ± 5.0		
$3s[3/2]2-4d[5/2]3$	$2\ 701.4 \pm 2.4$	2 469.4	232.0 ± 2.4		
$3s[3/2]2-4d[7/2]3$	$2\ 696.1 \pm 5.2$	2 465.8	230.3 ± 5.2		
$3s[3/2]2-4d[7/2]4$	$2\ 697.0 \pm 3.6$	2 465.7	231.3 ± 3.6		

Table V. — *Analysis of the experimental results for the $2p^5 5s$ configuration (all the data are in MHz).*

Two-photon transition	Isotopic shift $^{20}\text{Ne}-^{22}\text{Ne}$ experimental value Δ_{exp}	Bohr mass shift (calculated) Δ_B	Specific shift $\Delta_S = \Delta_{\text{exp}} - \Delta_B$	Shift of $J=1$ levels $\Delta_S(J=1) - \Delta_S(J \neq 1)$	Specific shift due to the subconfiguration $\overline{\Delta_S}(j_1=1/2) - \overline{\Delta_S}(j_1=3/2)$
$3s[3/2]2-5s'[1/2]0$	$2\ 646.8 \pm 3.6$	2 436.3	210.5 ± 3.6		} 26.4 ± 5.0
$3s[3/2]2-5s'[1/2]1$	$2\ 621.4 \pm 2.3$	2 440.0	181.4 ± 2.3	$- 29.1 \pm 5.9$	
$3s[3/2]2-5s[3/2]1$	$2\ 532.5 \pm 3.0$	2 384.4	148.1 ± 3.0	$- 36.0 \pm 5.4$	
$3s[3/2]2-5s[3/2]2$	$2\ 562.1 \pm 2.4$	2 378.0	184.1 ± 2.4		

(labelled Δ_{exp} first column). The values of the isotopic shift have been obtained for all the transitions except the $3s[3/2]2-4d[1/2]0$. In the latter case the two-photon transition probability (see table II) is too low and it has not been possible to observe the line corresponding to neon 22.

3. Theoretical analysis. — The isotopic shift is due to nuclear effects. There are two types of isotopic shifts : (i) the volume effect is related to the non Coulomb character of the electrostatic potential inside the nucleus (ii) the mass effect is related to the fact that the centre of mass of the atom does not correspond to the centre of the nucleus. If we consider two isotopes, both effects will modify the energy of the atom in a different way for each. Nevertheless, it is wellknown that for light elements such as neon the mass effect is much more important than the volume effect. (As a matter of fact, we did compare the ^{20}Ne - ^{21}Ne and ^{20}Ne - ^{22}Ne isotopic shifts, and no evidence of a volume effect was found [2].) So, we shall discuss the present experimental results in terms of the mass shift only.

The mass shift includes three terms : two of them can be derived very simply from non-relativistic quantum mechanics by adding the kinetic energy of the nucleus to the electronic hamiltonian ; as shown below, they give the Bohr shift and the main part of the specific shift. The third term which also contributes to the specific shift comes from relativistic corrections and second order contributions ; it will be dealt with at the end of the paragraph. Let us just point out for the moment that it depends only on the *core* angular momentum, which can take two values $1/2$ and $3/2$. So, within a configuration it brings about different shifts for the two sub-configurations corresponding to the two values of the core. This difference is about 25 MHz.

In non relativistic quantum mechanics, the mass-shift is related to a perturbation equal to $P^2/2M$ where \mathbf{P} is the momentum of the nucleus. In the frame where the total momentum of the atom is equal to 0, \mathbf{P} is equal to $-\sum_i \mathbf{p}_i$ where \mathbf{p}_i is the momentum of the i th electron. We thus obtain :

$$\frac{P^2}{2M} = \sum_i \frac{p_i^2}{2M} + \sum_{i \neq j} \frac{\mathbf{p}_i \cdot \mathbf{p}_j}{M}.$$

The first term gives the Bohr mass-shift while the second one is predominant in the calculation of the specific mass-shift [12, 13]. It can easily be shown that the Bohr mass-shift corresponds to the shift obtained when the mass of the electron m is replaced in the unperturbed Hamiltonian by the reduced mass $\mu = mM/(m + M)$ where M is the mass of the nucleus. The corresponding shift δE_e of an excited state whose energy is E_e is equal to

$$\delta E_e = - \frac{m}{m + M} E_e.$$

Since δE_e depends on M , two different isotopes have different level shifts which results in an isotopic shift for the transition. The value of the Bohr isotopic shift for each transition is reported in tables IV and V (labelled Δ_B). In the case of a one electron atom, the Bohr mass-shift is the only term of the mass shift. In the case of a many-electron atom, other contributions appear, the principal of which is the second term of the mass shift correction given before. The various terms which contribute to the specific mass-shift in neon have been analyzed by Bauche and Keller [14, 15]. For the present analysis, we can only retain two of them.

(a) The above mentioned operator $\sum \mathbf{p}_i \cdot \mathbf{p}_j / M$ gives rise on the one hand to an overall shift of each configuration and, on the other hand, within a configuration, to an additional shift of the $J = 1$ states only. This fact can be accounted for by considering the tensorial analogy between the angular part of the operator $\mathbf{p}_i \cdot \mathbf{p}_j$ and the operator $\mathbf{r}_i \cdot \mathbf{r}_j$ which appears in the expansion of the electrostatic interaction between two electrons. It is clear that the matrix elements of the first operator can be different from 0 only if the same property is true for the second. Because this last term is related to the $G^1(nl, n'l - 1)$ Slater integral, we deduce the following rule : the specific mass-shift may change from one Russel-Saunders term T_1 to another term T_2 in a pure configuration only if at least one G^1 Slater integral appears for the formal expression for the energy difference between T_1 and T_2 [12].

In the case of the $2p^5 4d$ and $2p^5 5s$ configurations, there is a G^1 Slater integral in the electrostatic energy difference of two Russel-Saunders terms only if one of them is a 1P term [20].

But the LS coupling is a rather bad approximation in these configurations and we have to use the intermediate coupling method [12, 13]. The experimental determination of the isotopic shift is thus a method of getting the expansion of the 1P term on the real eigenstates of the atom. More precisely, let us consider the difference between the specific shifts of a state having a component of 1P (state with $J = 1$) and of the other states of the same subconfiguration (1) having no component of 1P . In atomic units, this difference equals [12, 17] :

$$\Delta_S(\tau_1) - \bar{\Delta}_S(J \neq 1) = g_1 2m \left[\frac{1}{M_1} - \frac{1}{M_2} \right] \times (\tau_1 | ^1P)^2 J^2(2p, n\ell). \quad (1)$$

(¹) The five electrons of the almost closed $2p$ shell have an angular momentum which can take the values $j_1 = 1/2$ or $3/2$. In the $4d$ and $5s$ configuration, j_1 is almost a good quantum number, and the levels are arranged in two groups, or subconfigurations corresponding to the two values of j_1 . Because of relativistic effects which will be dealt with below, the specific shifts of the two sub-configurations of a configuration are different.

In this expression g_1 is the coefficient of the Slater integral G^1 in the expansion of the electrostatic interaction, its value is equal to $\frac{4}{3}$ in the case of the $2p^5 4d$ configuration and $\frac{2}{3}$ in the case of the $2p^5 5s$ [20]. m , M_1 and M_2 are respectively the mass of the electron and of the two isotopes of neon. $(\tau_1 | ^1P)^2$ is the square of the component of the wavefunction of the excited state $|\tau, J = 1\rangle$ on the 1P state. $J(2p, n\ell)$ is a radial integral, its value is equal to [21] :

$$J(n\ell, n' \ell - 1) \equiv J(n' \ell - 1, n\ell) = \int_0^{+\infty} R_{n\ell}(r) \times \left(\frac{d}{dr} - \frac{\ell - 1}{r} \right) R_{n' \ell - 1}(r) r^2 dr .$$

The analysis of our experimental results (tables IV and V) shows that within the experimental error, the specific shifts of the levels with $J \neq 1$ are actually the same in a given subconfiguration. Taking the difference between its mean value $\Delta_S(J \neq 1)$ and the isotopic shifts for the states with $J = 1$ gives an experimental determination of the left hand side of (1). As the sum over the $J = 1$ states of a configuration $\sum_{\tau_1} (\tau_1 | ^1P)^2$ equals 1, the sum over the same levels of $\Delta_S(\tau_1) - \bar{\Delta}_S$ is directly related to the radial integral $J^2(2p, n\ell)$ while the fractional values of $\Delta_S(\tau_1) - \bar{\Delta}_S$ give the expansion of the 1P state on the real eigenstates of the atom.

We thus deduced from the experimental determination of the isotopic shifts the quantities $(\tau_1 | ^1P)^2$. The experimental determinations are compared with theoretical predictions of Liberman [8] in tables VI and VII. As can be seen, the agreement is excellent.

Table VI. — Expansion of the 1P Russel-Saunders term over the real eigenstates of the atom for the $2p^5 4d$ configuration (value of $(\tau | ^1P)^2$).

	4d[3/2]1	4d[1/2]1	4d[3/2]1
Experiment	0.29	0.17	0.54
Theory	0.31	0.17	0.52

Table VII. — Expansion of the 1P Russel-Saunders term over the real eigenstates of the atom for the $2p^5 5s$ configuration (value of $(\tau | ^1P)^2$).

	5s[1/2]1	5s[3/2]1
Experiment	0.45	0.55
Theory	0.41	0.59

This allows us to give also the experimental determinations of the radial integrals $J^2(2p, 4d)$ and $J^2(2p, 5s)$ which are equal to :

$$J^2(2p, 4d) = 2.61 \pm 0.67 \times 10^{-3} \text{ atomic units ,}$$

$$J^2(2p, 5s) = 5.95 \pm 0.65 \times 10^{-3} \text{ atomic units .}$$

The values are also in very good agreement with theoretical predictions of Bauche [9]. The calculations of Bauche were performed in the Hartree-Fock scheme, using the computer code of Froese-Fischer [22]. He finds :

$$J_{th}^2(2p, 4d) = 2.67 \times 10^{-3} \text{ atomic units ,}$$

$$J_{th}^2(2p, 5s) = 4.93 \times 10^{-3} \text{ atomic units .}$$

The mean value Δ_S^m of the specific mass-shift of a configuration can be calculated by the same method. However the results in this case appear to be worse. For instance the theory predicts that Δ_S^m would be slightly larger for the transitions reaching the $2p^5 5s$ configuration than for the ones which attain $2p^5 4d$. In fact, it appears experimentally that the opposite behaviour prevails, $\Delta_S^m(2p^5 4d)$ being about 36 MHz larger than $\Delta_S^m(2p^5 5s)$. The comparison between the excellent results obtained within a configuration and the poorer results in the case of two different configurations yields an indication of the possibilities and limitations of the present theory.

(b) In addition, one must take into account relativistic corrections to the mass-shift [16, 17] which have the same angular dependence as the spin-orbit interaction. These corrections and second order perturbation terms involving spin-orbit interaction plus the mass-shift Hamiltonian [15] bring about a contribution to the isotopic shift which depends on the value of the angular momentum j_1 of the core. From the analysis of the measurements of Odintsov [18] and Keller and Lesprit [19], Keller [15] deduced that these terms lead to an isotopic shift ^{20}Ne - ^{22}Ne larger for the excited levels with $J_1 = 1/2$ than for those with $J_1 = 3/2$ by an amount Δ_R of the order of 25 MHz. Our result shows a good agreement with this value (see tables IV and V, last column).

Conclusion. — These experiments show that the measurements of isotopic shifts in a whole configuration permits one to obtain much valuable information on the wavefunctions of the excited state. It also shows that the improvements of the experimental set-up allows the application of the Doppler-free two-photon excitation methods (even by using c.w. dye laser) to numerous levels for which the transition probabilities are very small. It is clear that by using other dyes the present experiments can be extended to other configurations of neon and to other noble gases.

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