Study of $3d-4f$ and $3d-5f$ quartet and doublet transitions of doubly excited three-electron ions

Experiment and MCDF theory along the isoelectronic sequence

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We present the first observation of optical transitions between doubly excited doublet states in the term systems N V, O VI and F VII. The spectra were produced by foil excitation of fast ion beams. The assignment of the spectral lines was made by comparison with the results of MCDF calculations along the isoelectronic sequence. The same method also led to the identification of two $3d-4f$ quartet transitions in Mg X.

I. Introduction

Previous reports on doubly excited three-electron ions by the Bochum group [1, 2] dealt with transitions to $n=2$ levels in a number of ions. In the present work we report on similar measurements of $n=3-n'=4, 5$ transitions in N V, O VI, F VII and Mg X. N V has recently been reviewed by Garnir et al. [3]. We concentrate on transitions from the configurations $1s2s4f$, $1s2s5f$, $1s2p4f$ and $1s2p5f$, because these high angular momentum states are preferentially populated in the ion-foil interaction. Furtheron they cannot decay to $n=2$ states by optical electric dipole transitions. Figure 1 gives a schematic overview of the transitions in question. In [1] some tentative assignments of $n=3-n'=4$ transitions in O VI and F VII were presented, but at that time without any well-founded theoretical support. Meanwhile, Laughlin et al. [4] published the results of model-potential calculations of wavelengths and lifetimes for the core-excited $1s2snl$ ^4L and $1s2pnl$ ^4L levels of the ions O^{5+} and F^{6+}. We agree with general considerations put forward in that study concerning lines which are expected to be strong and which are therefore prime candidates for observation. Based on his wavelengths calculations, Laughlin et al. reassigned a few of the lines reported in [1]. We in turn, following Laughlin et al., could identify several $n=3-n'=5$ transitions in other spectra.

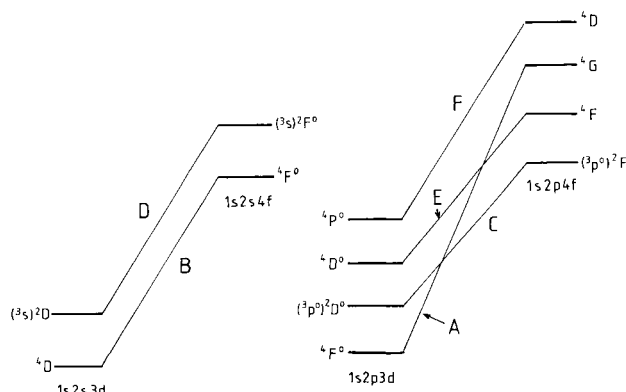However, a few lines that we knew to originate

Fig. 1. Term diagram of $n=3$ and $n'=4$ levels of doubly excited states, related to transitions reported. The level ordering is the one calculated for nitrogen. The transitions labeled A–F are discussed in the text and referred to in the tables

from three-electron states remained unidentified in our spectra. Therefore we performed MCDF-calculations (Multiconfiguration Dirac-Fock) using the Desclaux code [5] both to support our quartet assignments and to obtain theoretical values on doublet term levels. After determining the reliability of the calculations, several of our observed lines could be identified with transitions in the core-excited doublet-state systems. Extrapolating along the isoelectronic sequence the deviation between measurement and MCDF-approach results exact wavelengths in Mg X could be predicted and several lines be identified in this spectrum.

II. Experiment

The experiments were done in at the Bochum 4 MV Dynamitron Tandem accelerator laboratory. Whereas for the elements N, O, and F stable ion beam currents of several μA could be used, for Mg ions only currents of order 300 nA were available. The ions were excited by being passed through thin carbon foils (about 20 μg/cm^2) produced from a glow discharge. Spectra were taken using a 2.2 m grazing-incidence monochromator (McPherson Mod. 247) which viewed the ion beam at right angles. For these experiments we used a ruled grating of 600 l/mm. For wavelengths beyond 70 nm (as for N V) the efficiency of this grating was only just sufficient [13]. The linearity of the exit slit displacement was monitored by a 60 mm travel Heidenhain moiré fringe length gauge. A detailed description of the apparatus used can be taken from Träbert et al.[6].

III. Theory

For our theoretical calculations the latest version of the Desclaux code [5] was used. In our approach, we use all relativistic configurations coupled to good total angular momentum J which can be constructed from one electron wavefunctions which are usually denoted in the non relativistic way, like e.g. $1s2p3p$. Each of these relativistic configurations which in itself are linear combinations of Slater determinants build up the basis in the Multiconfiguration approach. Having evaluated all possible levels for all J, the levels are grouped in certain LS-terms averaging over the fine structure sublevels statistically in order to obtain LS-energy levels, because the fine structure is not resolved in the spectra. These self-consistent calculations also include in the energy the Breit-interaction i.e. the magnetic interaction and retardation plus a good approximation to the QED-effects in perturbation theory. There are some convergence problems for configurations of the type $1s2s3s$ or $1s2p4p$, but for levels which are of interest in this paper, e.g. $1s2p3d$ and $1s2s5f$, the calculations are in fact straightforward.

IV. Results and discussion

In general, $1s2lnf'$ states cannot decay to $n=2$ states directly by an electric dipole transition, and they are well populated as high angular momentum states by foil excitation of fast ion beams. As can be seen from Table 1 we expect five $n=3-n'=4$ quartet transitions to be strong (labeled A to E). These labels are used

Table 1. Transitions of the type $1s2l3dL - 1s2l\ 4/5fL'$. The labeled ones (A–F) are expected to provide lines strong enough to be observed in photon spectra. Relative intensities are calculated from:

$$(2L+1)(2L'+1)\left\{\begin{matrix} L_{core} & L & d \\ 1 & f & L' \end{matrix}\right\}^2$$

Transition		Label	Relative intensity
$1s2s3d\,^4D$	$-1s2snf\,^4F^0$	B	
$1s2s(^3S)3d\,^2D$	$-1s2s(^3S)nf\,^2F^0$	D	
$1s2p3d\,^4F^0$	$-1s2pnf\,^4G$	A	1.285
$^4F^0$	$-\quad\quad ^4F$		0.111
$^4D^0$	$-\quad\quad ^4F$	E	0.888
$^4F^0$	$-\quad\quad ^4D$		0.003
$^4D^0$	$-\quad\quad ^4D$		0.111
$^4P^0$	$-\quad\quad ^4D$	F	0.600
$1s2p(^3P^0)3d\,^2D^0$	$-1s2p(^3P^0)nf\,^2F$	C	

throughout this report and they are based on wavelength ordering.

Of these the $1s2s3d\,^4D - 1s2s4f\,^4F^0$ transition (Label B) is certainly strong and has been detected as the important cascade level appearing in decay curves of the $1s2s3d\,^4D$ level [7]. A rough calculation of the transition probabilities in LS-coupling demonstrates which of the possible transitions of the array $1s2p3d\,^4L^0 - 1s2pnf\,^4L'$ will be strong (Table 1). As expected we did not observe any of the $^4D^0 - ^4D$, $^4F^0 - ^4F$ or $^4F^0 - ^4D$ transitions for any of the elements studied. Concerning the doublet transitions, the $1s2s4f$ level cannot decay to a $1s^2$-core level by a single-electron transition, and the distance between the $2s$ and $4f$ electrons might be large enough to suppress rapid autoionization. Coulomb autoionization is in fact forbidden for the $1s2p4f\,^2F$ level because the $1s^2\,ef\,^2F^0$ term has the wrong parity. Therefore we believe that an optical transition from the $1s2p(^3P^0)4f\,^2F$ level, – in contrast to ^2D and ^2G –, to $1s2p(^3P^0)3d\,^2D^0$ should be observable. Singlet-core states of this configuration decay preferentially to $1s^2nl\,^2L$.

In Tables 2, 3 and 4 we present our experimental results on the various transitions comparing the data with those from other experiments and from various theoretical approaches.

The calculations by Laughlin [4] helped with the identifications of a number of quartet transitions. With this aid also several $3d-4f$ and $3d-5f$ transitions in O VI and F VII are identified. One should notice that in most cases our measured wavelengths are shorter than Laughlin's values. In order to identify $1s2p3d\,^4D^0 - 1s2p5f\,^4F$ (Label E) and $1s2p3d\,^4P^0 - 1s2p5f\,^4D$ (Label F) we used a method described below. For N V Garnir et al. [3] give results for the $3d-4f$ quartet transitions in comparison with

Table 2. Wavelengths of transitions in N^{4+}

Label	Transition		Experiment			Theory	
			This work λ/nm	Relative intensity	Other experiments [3] λ/nm	MCDF This work λ/nm	Other [3] λ/nm
$n=3-n'=4$							
A	$1s2p3d\,^4F^0$	$-1s2p4f\,^4G$	67.294 ± 0.007	55	63.318 ± 0.005	67.475	
B	$1s2s3d\,^4D$	$-1s2s4f\,^4F^0$	67.375 ± 0.01	60	67.390 ± 0.005	69.587	67.300
C	$1s2p(^3P^0)3d\,^2D^0$	$-1s2p(^3P^0)4f\,^2F$	68.694 ± 0.01	10		69.057	
D	$1s2s(^3S)3d\,^2D$	$-1s2s(^3S)4f\,^2D^0$	71.273 ± 0.008[a]	12		73.919	
E	$1s2p3d\,^4D^0$	$-1s2p4f\,^4F$	72.489 ± 0.005	25	72.498 ± 0.006	72.802	72.517
F	$1s2p3d\,^4P^0$	$-1s2p4f\,^4D$	73.753 ± 0.02	30	73.77 ± 0.01	73.054	73.801
$n=3-n'=5$							
A	$1s2p3d\,^4F^0$	$-1s2p5f\,^4G$	47.310 ± 0.02	10		47.659	
B	$1s2s3d\,^4D$	$-1s2s5f\,^4F^0$	47.503 ± 0.02	15		48.651	
E	$1s2p3d\,^4D^0$	$-1s2p5f\,^4F$	49.875 ± 0.02	12		50.039	

[a] Tentative assignment

Table 3. Wavelengths of transitions in O^{5+}

Label	Transition		Experiment		Theory	
			This work λ/nm	Relative intensity	MCDF This work λ/nm	Other [4] λ/nm
$n=3-n'=4$						
A	$1s2p3d\,^4F^0$	$-1s2p4f\,^4G$	47.285 ± 0.005	170	47.454	47.359
B	$1s2s3d\,^4D$	$-1s2s4f\,^4F^0$	47.451 ± 0.005	270	48.695	47.459
C	$1s2p(^3P^0)3d\,^2D^0$	$-1s2p(^3P^0)3f\,^2F$	48.299 ± 0.009	120	48.459	
D	$1s2s(^3S)3d\,^2D$	$-1s2s(^3S)4f\,^2F^0$	49.989 ± 0.008[a]	80	51.333	
E	$1s2p3d\,^4D^0$	$-1s2p4f\,^4F$	50.608 ± 0.01	150	50.752	50.653
F	$1s2p3d\,^4P^0$	$-1s2p4f\,^4D$	51.262 ± 0.008	140	50.796	51.395
$n=3-n'=5$						
A	$1s2p3d\,^4F^0$	$-1s2p5f\,^4G$	33.175 ± 0.007	110	33.388	33.212
B	$1s2s3d\,^4D$	$-1s2s5f\,^4F^0$	33.348 ± 0.006	120	33.965	33.355
C	$1s2p(^3P^0)3d\,^2D^0$	$-1s2p(^3P^0)5f\,^2F$	34.667 ± 0.01	30		
E	$1s2p3d\,^4D^0$	$-1s2p5f\,^4F$	34.75 ± 0.02	120	34.860	34.798
F	$1s2p3d\,^4P^0$	$-1s2p5f\,^4D$	35.332 ± 0.015	40	35.127	35.405

[a] Tentative assignment

Chung's theoretical values [3]. We agree with their analysis for $1s2s3d\,^4D-1s2s4f\,^4F^0$, $1s2p3d\,^4D^0-1s2p4f\,^4F$, and $1s2p3d\,^4P^0-1s2p4f\,^4D$. However, we believe that the line they reported at $\lambda=(67.288\pm0.004)$ nm and we measured at $\lambda=(67.294\pm0.007)$ nm should be classified as $1s2p3d\,^4F^0-1s2p4f\,^4G$ instead of $1s2p3d\,^4F^0-1s2p4f\,^4F$. The reasons are obvious from Table 1 and from considering relative intensities given in Tables 2, 3 and 4. By the way, Chung's calculations [3] did not cover the ^4G level.

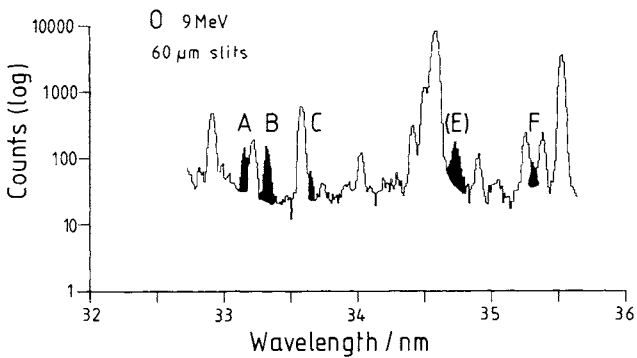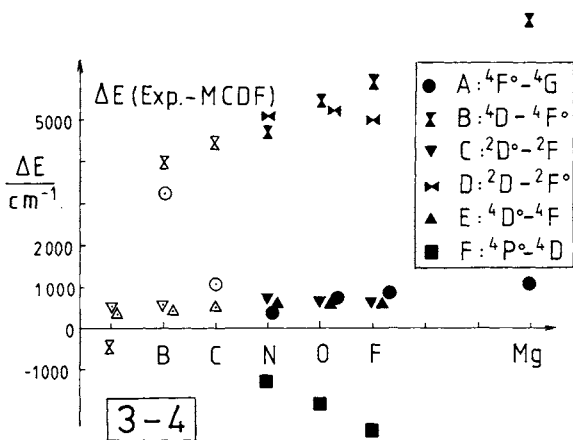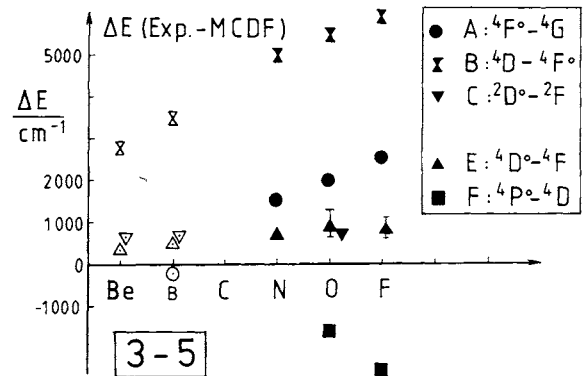At this point we started our own calculations using the MCDF-code [5]. At a first view the computed wavelength data are poor but we used the following method to get a strong support from these calculations for our assignments and help for further identifications: For each transition the transition energy was evaluated both from the experimental wavelengths and the MCDF results. The difference between experiment and theory was then plotted versus Z (Data can be found in Table 6.). This was done for $n=3-n'=4$ and $n=3-n'=5$ transitions as far as experimental data were available, including our own assignments, as presented in this report (Figs.3 and 4). From these graphic representations one can see that in most cases the deviation between experiment and theory is rather linear in Z, or even approximately constant over Z for some transitions.

Table 4. Wavelengths of transitions in F^{6+}

Label	Transition		Experiment		Theory	
			This work λ/nm	Relative intensity	MCDF This work λ/nm	Other [4] λ/nm
$n=3-n'=4$						
A	$1s2p3d\,^4F^0$	$-1s2p4f\,^4G$	35.106 ± 0.005	290	35.216	35.155
B	$1s2s3d\,^4D$	$-1s2s4f\,^4F^0$	35.258 ± 0.005	290	36.005	35.270
C	$1s2p(^3P^0)3d\,^2D^0$	$-1s2p(^3P^0)4f\,^2F$	35.808 ± 0.005	150	35.896	
D	$1s2s(^3S)3d\,^2D$	$-1s2s(^3S)4f\,^2F^0$	36.920 ± 0.007^a	125	37.616	
E	$1s2p3d\,^4D^0$	$-1s2p4f\,^4F$	$37.32\ \pm0.02$ bl	130	37.401	37.340
F	$1s2p3d\,^4P^0$	$-1s2p4f\,^4D$	37.695 ± 0.005	230	37.371	37.695
$n=3-n'=5$						
A	$1s2p3d\,^4F^0$	$-1s2p5f\,^4G$	24.546 ± 0.005	50	24.695	24.564
B	$1s2s3d\,^4D$	$-1s2s5f\,^4F^0$	24.702 ± 0.005	60	25.065	24.702
E	$1s2p3d\,^4D^0$	$-1s2p5f\,^4F$	25.634 ± 0.01	40	25.675	
F	$1s2p3d\,^4P^0$	$-1s2p5f\,^4D$	25.963 ± 0.01	30	25.778	

^{a} Tentative assignment
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Fig. 2. Beam-foil spectrum of oxygen, recorded with a 2.2 m grazing-incidence spectrometer. $N=3-n'=5$ transitions between core-excited states are marked in black and labeled as in Table 3

Fig. 4. Same as in Fig. 3, but data refer to $n=3-n'=5$ transitions

Fig. 3. Deviation between experiment and MCDF-approach of the transition energy of some $n=3-n'=4$ transitions. If not marked otherwise, error bars are smaller than symbols. Full symbols indicate present observations

This can be understood in the following way. In our theoretical approach we exclude important parts of the correlation energy as we allow only for mixing of states that belong to a single non-relativistic configuration. This neglected correlation energy can to first order be either linear in Z or constant. As we learned from Brage's work [12], it is linear, if other configurations which belong to the same so-called complex possess the same LS-term [13]. A complex consists of configurations that have the same quantum numbers n in all electrons. Consider now the energy shift due to mixing in second order perturbation theory from inside and outside the complex:

$$\Delta E_2 = \frac{\langle H_{ij}\rangle^2}{E_i - E_j}.$$

In all cases the matrix elements $\langle H_{ij}\rangle$ are linear in Z as they are proportional to Slater-integrals. However for mixing inside a complex, $E_i - E_j$ is linear

in Z, making ΔE_2 linear, too. On the other hand, for mixing from outside the complex $E_i - E_j$ is proportional to Z^2 and therefore ΔE_2 is independent of Z.

From this follows that the deviation between experimental and theoretical transition energies will be constant over Z if the two energy levels that belong to a transition are the only members in their complex. As an example, see the transition $1s2p3d\,^4F^0 - 1s2p4f\,^4G$. There is no other $n=3\,^4F^0$ term and there is no other $n=4\,^4G$ term either. The observed deviation is in fact constant (Fig. 3). On the other hand, considering the transition $1s2p3d\,^4F^0 - 1s2p5f\,^4G$, there is the term $1s2s5g\,^4G$ in the same complex as the $1s2p5f\,^4G$ term, pushing up $1s2p5f\,^4G$ in energy. Therefore, taking this mixing into account, the experimental transition energy in fact gets larger than our calculated value. Without this correction the deviation should be linear in Z as can actually be seen from Fig. 4.

Checking every transition carefully, our plotted deviations follow in each case these simple arguments of linearity. When we noticed this, we extrapolated the observed deviations between experiment and theory to higher values of Z. For the case of Mg X, for which we have experimental data, the $1s2s3d\,^4D - 1s2s4f\,^4G^0$ and $1s2p3d\,^4F^0 - 1s2p4f\,^4G$ transitions are expected to be strong. The above extrapolation yielded a good prediction of the wavelength range where to look for these transitions in a Mg spectrum (Table 7). In fact, we observed two features that can be identified with these transitions (Fig. 5). The deviations calculated from these two transitions are included in Fig. 2.

Our plots give us confidence about the identification of the doublet transitions $1s2p(^3P^0)3d\,^2D^0 - 1s2p(^3P^0)4f\,^2F$. The deviation data for $1s2s(^3S)3d\,^2D - 1s2s(^3S)4f\,^2F^0$ are not close enough to the aforementioned trends so that we consider our assignments as being more than tentative. Anyhow, the identification of $1s2p(^3P^0)3d\,^2D^0 - 1s2p(^3P^0)5f\,^2F$ in O VI (see Fig. 2) appears as being realistic. In the spectra of N and F this transition has not been observed, since at that time we did not expect these doublet lines to be visible, and thus we recorded these wavelength regions with insufficiently long integration times.

For nitrogen we could not observe $1s2p3d\,^4P^0 - 1s2p5f\,^4D$. Generally the transitions $^4P^0 - ^4D$ are weak as can be seen from the tables. We did not try to find analogous transitions in C IV as they are known to be in a wavelength region where the efficiency of our spectrometer is not sufficient to permit a proper experiment [13].

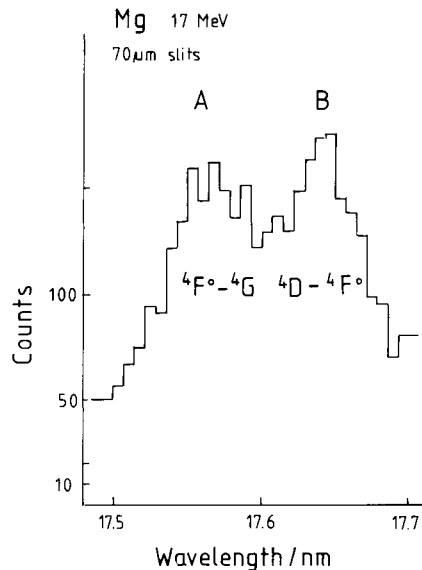For lower members of the isoelectronic sequence we also evaluated deviation points whereever experi-

Fig. 5. Part of a beam-foil spectrum recorded using 17 MeV Mg-ions. The transitions Mg X $1s2p3d\,^4F^0 - 1s2p4f\,^4G$ and $1s2s3d\,^4D - 1s2s4f\,^4F^0$ appear. In between these lines there is an indication of the Mg X $1s^22s\,^2S - 1s^24p\,^2P^0$ line appearing in fourth diffraction order

Table 5. Experimental wavelengths used for data in Table 6. For interpretation of label, see Table 1

Transition label	Be [8, 9] λ/nm	B [10] λ/nm	C [11] λ/nm
$n=3-n'=4$			
A		170.56 ± 0.01	102.57 ± 0.02
B	432.955 ± 0.007	176.49 ± 0.01	102.89 ± 0.02
C	399.55 ± 0.03	183.68 ± 0.01	
D			
E	437.11 ± 0.01	197.9 ± 0.02	112.53 ± 0.02
F			
$n=3-n'=5$			
A		125.12 ± 0.01	
B	276.4 ± 0.1	127.56 ± 0.01	
C	284.53 ± 0.03	129.68 ± 0.02	
D			
E	303.1 ± 0.1	136.63 ± 0.02	
F			

mental figures were known (Tables 5 and 6). In some cases the linear trend is apparent but there also exist cases with pronounced irregularities. Those might be due to mixing with Rydberg term series with a different electron-core and different principal quantum number n, an effect which is known to be important for low Z [8]. This effect occurs preferentially for just one member of the isoelectronic sequence. For example, the experimental data for B III $1s2p3d\,^4F^0 - 1s2p4f\,^4G$ result in a deviation point far away from expectation. Such deviations are caused by so called short-range interactions, whereas our ap-

Table 6. Deviation of transition energies between experimental data and MCDF-approach results. For visualisation the same data are plotted in Figs. 3 and 4

Transition label	Be $\Delta E/\mathrm{cm}^{-1}$	B $\Delta E/\mathrm{cm}^{-1}$	C $\Delta E/\mathrm{cm}^{-1}$	N $\Delta E/\mathrm{cm}^{-1}$	O $\Delta E/\mathrm{cm}^{-1}$	F $\Delta E/\mathrm{cm}^{-1}$	Mg $\Delta E/\mathrm{cm}^{-1}$
$n=3-n'=4$							
A		3283 ± 3	1094 ± 20	398 ± 25	753 ± 20	890 ± 40	1132 ± 160
B	-399 ± 1	3987 ± 3	4403 ± 20	4718 ± 25	5383 ± 20	5880 ± 40	7411 ± 170
C	557 ± 3	593 ± 3		765 ± 25	683 ± 40	685 ± 40	
D				5022 ± 20	5237 ± 40	5012 ± 60	
E	343 ± 2	422 ± 5	552 ± 20	593 ± 15	561 ± 40	580 ± 150	
F				-1279 ± 45	-1790 ± 35	-2300 ± 80	
$n=3-n'=5$							
A	-205 ± 6			1547 ± 100	1923 ± 70	2458 ± 70	
B	2798 ± 15	3465 ± 6		4967 ± 100	5447 ± 60	5862 ± 80	
C	615 ± 3	642 ± 15					
D							
E	375 ± 15	500 ± 15		657 ± 100	908 ± 300	623 ± 150	
F					-1657 ± 100	-2764 ± 150	

Table 7. Wavelengths of transitions in Mg^{9+}. See spectrum in Fig. 5

Transition	MCDF ab initio λ/nm	MCDF extrapolated λ/nm	Experiment λ/nm
$1s2p3d\,^4F^0-1s2p4f\,^4G$	17.603	17.560 ± 0.012	17.568 ± 0.005
$1s2s3d\,^4D-1s2s4f\,^4F^0$	17.886	17.657 ± 0.006	17.652 ± 0.005

proach can at most cope with long range-interactions that are important for all members of an isoelectronic sequence [14, 15].

Very recently Bouchama et al. [16] presented a method to produce spectra, in which quartet lines were enhanced (preferentially from upper levels of ions with a $1s2s$ core). We did not observe any of their $n=3-n'=4$ transitions, however. At $\lambda=(55.54\pm 0.01)$ nm Bouchama et al. identified a line as $1s2s3s\,^4S-1s2s4p\,^4P^0$. There is also a line in our survey spectra, which we at that time identified as $1s2p\,^1P^0-1s3d\,^1D$ in third order, according to our excitation method and the chosen ion energy of 3.5 MeV. This line has been reproduced with better resolution by Garnir et al. [3]. Our assignment does not exclude the assignment by Bouchama et al., since the third diffraction order line and the first order line might constitute a blend. Bouchama assigned a line at (73.02 ± 0.05) nm to NV $1s2s3d\,^4D-1s2s4p\,^4P^0$. In our spectra there is no line being present at this wavelength. On the other hand, Bouchama et al. present in their Fig. 3 a spectrum which covers the wavelength range where we identified N V $n=3-n'=4$ lines of core excited states. However none of these can be found in their spectrum, not even $1s2s3d\,^4D-1s2s4f\,^4F^0$, although this is a

$1s2s(^3S)$-core transition, that is of the type they claim to be excited preferentially. Nevertheless one should mention that they readily observed $1s2s2p\,^4P^0-1s2s4d\,^4D$, as we did by the beam-foil method [1]. However, the excitation processes seem not to be well understood for both light sources, presently.

In conclusion we were able to identify a number of $n=3-n'=4$ and $n=3-n'=5$ transitions in the term systems of N V, O VI, and F VII. Considering the validity limits of our MCDF-approach and making use of simple systematic trends the first observation between core excited doublet states in N V, O VI, and F VII could be manifested. Furthermore, by using our systematics for extrapolation we found two lines belonging to $n=3-n'=4$ quartet transitions in Mg X. We expect that by this method further quartet and doublet transitions can be predicted and will then be readily observed.

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