Coulomb capture and x-ray cascades of muons in metal halides

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Muonic x-ray yields have been measured for Al, AlF_3, $AlCl_3$, AlI_3, KCl, KBr, KI, Cd, CdF_2, $CdCl_2$, $CdBr_2$, CdI_2, Sn, $SnCl_2$, $SnCl_4$, Sb, SbF_3, $SbCl_3$, and $SbCl_5$. The yields were normalized with the total intensity of the Lyman series. The capture ratios between Coulomb capture in the anion and the cation were determined for all compounds listed. These ratios are not well reproduced by the Fermi-Teller "Z law." Chemical effects in the intensities were observed. All observed intensities were compared with cascade calculations assuming a modified statistical population of the levels with $n = 20$. In a number of cases this procedure does not yield a satisfactory reproduction of the experimental data.

I. INTRODUCTION

Although mesonic atoms have been known to exist for 28 years, little experimental work has been devoted to the problems of intensities and distribution of the mesonic particles between various components of a chemical compound. However, chemical effects were definitely established.[1-4] Coulomb capture, i.e., the transition of a particle from a continuous state to the first bound state, was not directly observed.

In a number of theoretical papers[5-17] Coulomb capture and subsequent Auger- and x-ray cascades were treated. The calculations are able to reproduce the observed x-ray intensities fairly well in many cases if a certain "initial population" with mesonic particles is assumed, i.e., a population of atomic bound states where the cascade calculation starts. However, no really satisfactory calculation of this initial distribution was achieved, at least in the case of condensed matter.

In this paper we present the results of experiments with muons. Comparison with theory will be made both for the ratio $A(Z'/Z)$ of the per-atom capture probabilities for capture in the constituents Z' and Z of a binary chemical compound Z'_kZ_m, and for the cascades. Chemical effects will be emphasized.

II. EXPERIMENT AND EVALUATION

The experiment was performed at the μ channel of the CERN synchrocyclotron. Setup and method were standard and have already been described.[18] Two Ge(Li) detectors were used: (1) planar, 5.4 cm^3, typical resolution 1.4 keV at 100 keV, useful energy range 150–1500 keV, or (2) coaxial, 18.5 cm^3, typical resolution 7 keV at 1000 keV, useful energy range 300–7000 keV. The detector efficiency was determined with calibrated radioactive sources, including the efficiency for off-axis source positions.

The target materials used in this experiment are listed in Table I. All targets, except $SnCl_4$ and $SbCl_5$ which were liquid, were solid, and at room temperature. The targets were 10×14 cm^2, and were tilted at 45° to the beam axis. The target thickness was 2.8 g/cm^2, and the effective thickness seen by the beam was 4 g/cm^2.

As examples, Figs. 1 and 2 display the high- and low-energy parts of the SbF_3 spectrum, respectively, and Fig. 3 the low-energy part of the AlF_3 spectrum. Spectra from the empty target boxes were also taken.

The spectra were evaluated and corrected for detector efficiency and for self-absorption with computer programs[19,20] developed by the CERN-Heidelberg-Karlsruhe-Munich group. Besides the

TABLE I. Targets and relative capture ratios $A(Z'/Z)$; the single-element targets of Al, Cd, Sn, and Sb are not listed in column 1.

Compound Z'_kZ_m	$A(Z'/Z)$ (expt.)	$A(Z'/Z)$ ("Z law"[5])	$A(Z'/Z)$ (theory[17])
AlF_3	0.90 ± 0.30	1.44	1.38
$AlCl_3$	0.63 ± 0.21	0.75	0.81
AlI_3	0.48 ± 0.18	0.24	0.37
KCl	1.16 ± 0.11	1.12	1.09
KBr	0.42 ± 0.07	0.54	0.65
KI	0.50 ± 0.08	0.36	0.50
CdF_2	3.98 ± 0.54	5.34	3.53
$CdCl_2$	2.26 ± 0.26	2.82	2.06
$CdBr_2$	0.95 ± 0.12	1.38	1.23
CdI_2	1.00 ± 0.12	0.90	0.94
$SnCl_2$	1.98 ± 0.22	2.94	2.11
$SnCl_4$ (liquid)	2.36 ± 0.40	2.94	2.11
SbF_3	3.69 ± 0.42	5.67	3.67
$SbCl_3$	2.55 ± 0.39	3.00	2.14
$SbCl_5$ (liquid)	2.30 ± 0.45	3.00	2.14

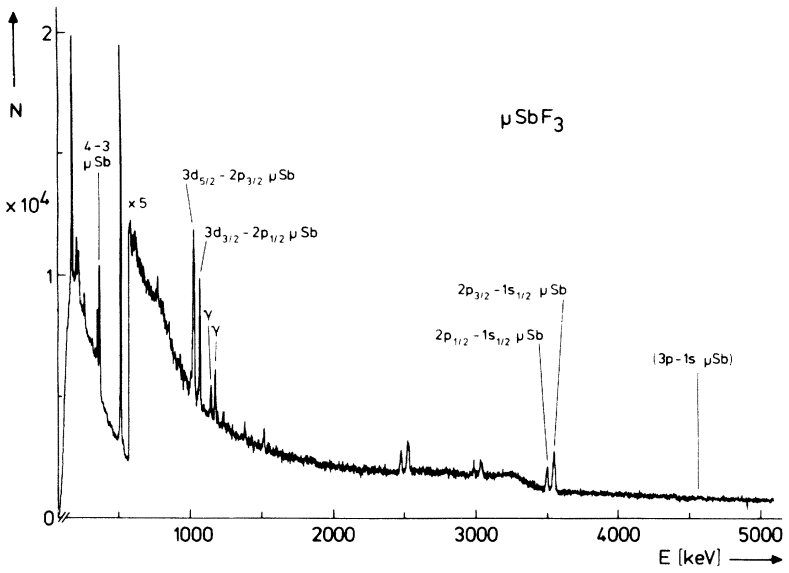

FIG. 1. Muonic x-ray spectrum of SbF_3 obtained with the coaxial diode.

statistical error of each line the following errors were taken into account: (1) target geometry, self-absorption, and detector efficiency, totaling 5%; and (2) normalization error (cf. Sec. III).

III. EXPERIMENTAL RESULTS

The experimentally determined intensities are summarized in Tables II–X. The first columns list the transitions characterized by the quantum numbers of upper and lower levels in the element given in the head of the column; for α and χ^2_ν, cf. Sec. IV. The other columns list the respective values for intensity and its total error for targets of the chemical form stated in the head of each column. The intensities are normalized to unity for the Lyman series which is justified for all the investigated elements.[21] Because of a lack of efficiency at higher energies only the $2p$-$1s$ line was seen in several cases (cf. Tables VI–X). In the case of Br its intensity was assumed to be $85 \pm 10\%$ of the total Lyman series, and in the cases of Cd, Sn, Sb, and I, to be $95 \pm 5\%$. These values are justified by recent experiments[22,23] and by cascade calculations of the present work. In some cases the $2p$-$1s$ lines were not measured, owing to lack

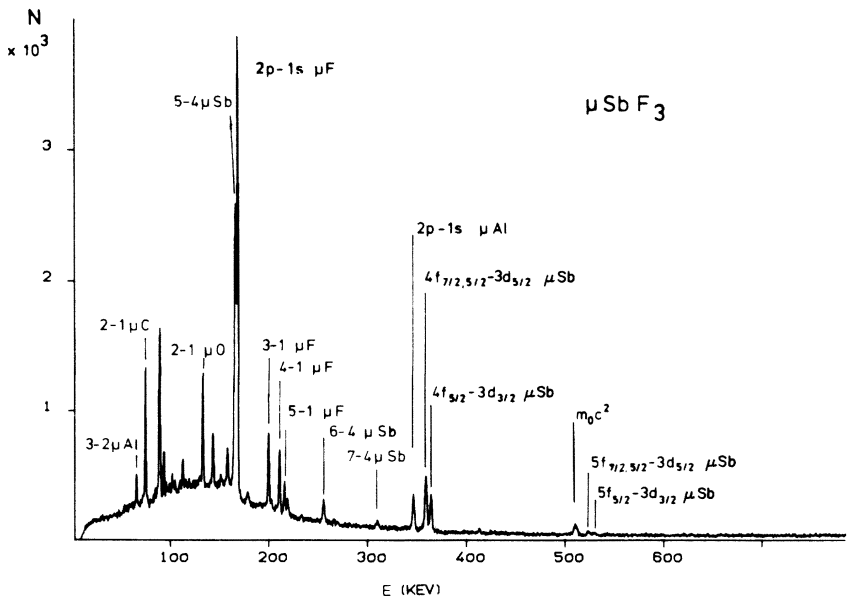

FIG. 2. Muonic x-ray spectrum of SbF_3 obtained with the planar diode.

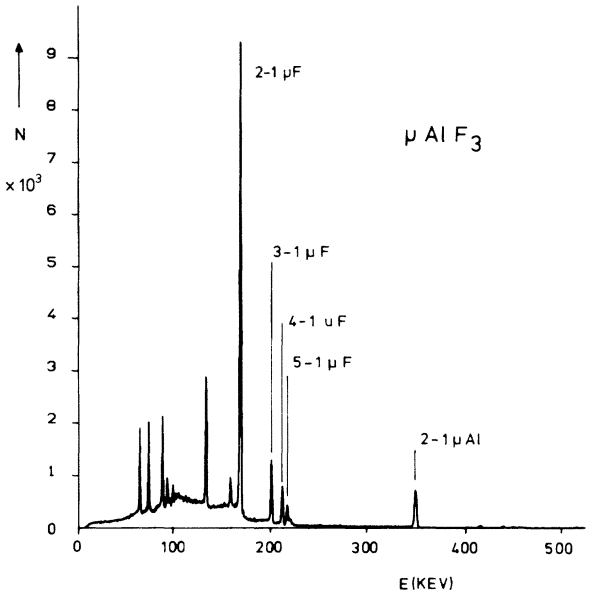

Fig. 3. Muonic x-ray spectrum of AlF_3 obtained with the planar diode.

of beam time (cf. Tables VI–X). In the cases of AlI_3, KBr, KI, CdF_2, and $CdCl_2$, we used for the normalization the measured $3d$-$2p$ intensities for Cd, Br, and I, and calculated the $2p$-$1s$ intensities by using the $(3d\text{-}2p)/2p\text{-}1s)$ intensity ratios as measured in $CdBr_2$ and CdI_2 for Br and I, respectively, and as measured in $CdBr_2$ for Cd. The assumptions on the $2p$-$1s$ intensities are in these cases the same as mentioned above.

From the experimental intensities muon distribution ratios $A(Z'/Z)$ were calculated with the formula

$$A(Z'/Z) = m\sum_n I_n(Z') \Big/ k\sum_n I_n(Z) ,$$

where $\sum_n I_n(Z')$ and $\sum_n I_n(Z)$ are the unnormalized intensities of the Lyman series in elements Z and Z, respectively. The distribution ratios are listed in column 2 of Table I.

Pronounced chemical effects are summarized in Table XI. These effects are given in form of double ratios because all errors owing to detector efficiency and normalization and most of the error owing to self-absorption cancel in this way. Moreover, the deviation of the listed numbers from unity is a direct measure of the chemical effect.

IV. CASCADE CALCULATIONS

In order to show systematic trends cascade calculations were performed[24] assuming an initial population at $n = 20$ proportional to

$$(2l+1)e^{\alpha l} ,$$

where l is the orbital angular momentum quantum number and α is a parameter. The choice of this n value is somewhat arbitrary. Experimental evidence has been reported,[25] however, that the calculations so performed reproduce the experimental results rather well.

The parameter α was varied in steps of 0.10. Each computed x-ray spectrum was compared with the corresponding experimental spectrum and the respective normalized χ^2_ν value[26] was determined (a good fit is characterized by $\chi^2_\nu \lesssim 1$). The α range was chosen so that the minimum was included. Tables II–X list the α values for the lowest χ^2_ν values obtained, and these χ^2_ν values.

V. DISCUSSION

For two of the distribution ratios $A(Z'/Z)$ measured in this work, comparison with earlier experimental work is possible. Zinov *et al.*[4] obtained $A(K/I) = 0.50 \pm 0.05$ and $A(Cd/I) = 1.00 \pm 0.20$, and

TABLE II. Intensities of muonic x-ray lines in F in several metal halides (in %).

F	AlF_3	CdF_2	SbF_3
7-1	0.3 ± 0.2		
6-1	1.4 ± 0.2		3.0 ± 0.4
5-1	4.3 ± 0.3	8.2 ± 1.3	6.2 ± 0.5
4-1	8.5 ± 0.6	10.8 ± 1.2	12.3 ± 0.8
3-1	12.5 ± 0.8	17.0 ± 1.5	15.0 ± 0.9
2-1	73.0 ± 1.2	64.0 ± 2.1	63.5 ± 1.5
α	0.2	0.1	0.1
χ^2_ν	4.5	2.6	5.3

TABLE III. Intensities of muonic x-ray lines in Al in several metal halides and pure metal (in %).

Al	Al metal	AlF_3	$AlCl_3$	AlI_3
8-1	0.6 ± 0.2			
7-1	1.0 ± 0.2	1.3 ± 1.1	1.7 ± 2.5	
6-1	2.6 ± 0.3	2.3 ± 0.9	3.8 ± 1.8	
5-1	4.1 ± 0.4	3.2 ± 1.0	2.4 ± 1.9	
4-1	5.4 ± 0.5	3.5 ± 1.1	4.2 ± 2.0	2.1 ± 6.3
3-1	7.7 ± 0.6	8.0 ± 1.4	10.3 ± 2.3	8.0 ± 6.8
2-1	78.6 ± 1.1	81.7 ± 2.5	77.5 ± 4.3	89.9 ± 9.6
α	0.2	0.3	0.2	
χ^2_ν	3.7	1.0	0.6	

TABLE IV. Intensities of muonic x-ray lines in Cl in several metal halides (in %).

Cl	$AlCl_3$	KCl	$CdCl_2$	$SnCl_2$	$SnCl_4$	$SbCl_3$	$SbCl_5$
7-1	1.9 ± 0.5						
6-1	1.5 ± 0.3	2.4 ± 1.0		2.2 ± 1.5			
5-1	2.8 ± 0.4	1.3 ± 0.9		4.9 ± 1.2	5.4 ± 1.9	3.5 ± 1.3	
4-1	2.7 ± 0.4	2.1 ± 1.1	2.6 ± 1.6	3.0 ± 1.1	3.5 ± 1.7	1.7 ± 1.1	3.0 ± 1.6
3-1	7.0 ± 0.6	9.8 ± 2.4	13.1 ± 1.7	7.4 ± 1.2	7.1 ± 1.9	6.3 ± 1.3	7.4 ± 1.6
2-1	84.1 ± 1.1	84.4 ± 2.8	84.3 ± 2.2	82.5 ± 2.3	84.0 ± 2.9	88.6 ± 2.5	89.6 ± 2.2
7-2	1.2 ± 0.3		1.0 ± 0.5	1.4 ± 0.5		1.4 ± 0.6	
6-2	2.7 ± 0.5	2.4 ± 0.5	3.7 ± 0.8	2.2 ± 0.5		2.6 ± 0.8	
5-2	4.6 ± 0.6	4.3 ± 0.9	5.3 ± 0.9	5.3 ± 0.9		4.6 ± 0.8	
α	0.2	0.2	0.2	0.1		0.2	
χ^2_ν	1.7	1.0	4.1	1.2		1.1	

Mausner *et al.*[27] found $A(K/Cl) = 1.16 \pm 0.03$, all in excellent agreement with our results.

Inspection of Table I shows that many values of our experimental distribution ratio $A(Z'/Z)$ do not agree with the "Z law."[5] There is, however, good agreement with a recent calculation by Daniel.[17] In this calculation the muon is classically treated as a particle losing energy in the electron Fermi gas. The muon is captured when the total (kinetic plus potential) energy becomes negative. The capture ratio per atom turns out to be

$$A(Z'/Z) = \frac{Z'^{1/3} \ln(0.57Z)}{Z^{1/3} \ln(0.57Z)} .$$

This formula was used to calculate the values in column 4 of Table I.

TABLE V. Intensities of muonic x-ray lines in K in several metal halides (in %).

K	KCl	KBr	KI
6-1	3.8 ± 1.5		
5-1	3.9 ± 1.6	2.8 ± 2.2	
4-1	2.9 ± 1.3	1.0 ± 1.9	
3-1	7.6 ± 1.9	5.1 ± 2.2	11.0 ± 4.0
2-1	81.8 ± 2.9	91.1 ± 3.4	89.0 ± 4.0
8-2		1.8 ± 0.7	
7-2	1.0 ± 0.3	0.8 ± 0.6	
6-2	1.9 ± 0.4	2.3 ± 0.7	3.6 ± 1.2
5-2	3.4 ± 0.5	4.4 ± 0.8	5.8 ± 1.5
4-2	9.4 ± 1.1	11.9 ± 1.7	6.5 ± 2.4
α	0.2	0.2	0.2
χ^2_ν	2.3	1.1	1.7

The measured x-ray intensities of Tables II–X show a great number of differences from target form to target form for a given element. Most of these differences are too small to be really significant when being considered isolated. However, some large chemical effects, as summarized separately in Table XI, were definitely established. Of course, it is far too early to try to understand all this data.

The comparison between experimental intensities and cascade calculations (Tables II–X) shows that there are a number of cases where the assumed modified statistical initial population, even for the adapted value of α, does not satisfactorily

TABLE VI. Intensities of muonic x-ray lines in Br in several metal halides (in %).

Br	KBr	CdBr
$2p_{3/2}$-$1s_{1/2}$		47.7 ± 6.7
$2p_{1/2}$-$1s_{1/2}$		37.3 ± 5.4
$4d_{3/2}$-$2p_{1/2}$		
$4d_{5/2}$-$2p_{3/2}$		
$3d_{3/2}$-$2p_{1/2}$	17.1 ± 2.6	17.9 ± 2.5
$3d_{5/2}$-$2p_{3/2}$	32.6 ± 5.4	31.5 ± 4.3
7-3	2.3 ± 0.7	
6-3	2.8 ± 0.6	2.8 ± 0.6
5-3	6.3 ± 1.1	6.6 ± 1.1
4-3	38.5 ± 6.3	40.3 ± 6.5
9-4	0.3 ± 0.4	
8-4	0.6 ± 0.3	
7-4	1.5 ± 0.4	
6-4	5.8 ± 1.0	4.7 ± 0.9
α	−0.2	−0.2
χ^2_ν	8.3	4.0

TABLE VII. Intensities of muonic x-ray lines in Cd in several metal halides and pure metals (in %).

Cd	Cd	CdF_2	$CdCl_2$	$CdBr_2$	CdI_2
$2p_{3/2}$-$1s_{1/2}$				62.0 ± 10.3	63.4 ± 15.9
$2p_{1/2}$-$1s_{1/2}$				33.0 ± 6.2	31.6 ± 9.3
$3d_{3/2}$-$2p_{1/2}$	22.4 ± 2.8	22.7 ± 4.4	23.0 ± 3.6	22.1 ± 3.7	22.8 ± 5.6
$3d_{5/2}$-$2p_{3/2}$	42.7 ± 4.9	42.4 ± 7.2	42.0 ± 5.9	42.6 ± 6.8	45.2 ± 10.6
$6f_{5/2}$-$3d_{3/2}$	1.7 ± 0.5		3.1 ± 2.3		
$6f_{7/2}$-$3d_{5/2}$	2.1 ± 0.6		3.7 ± 2.4		
$5f_{5/2}$-$3d_{3/2}$	2.7 ± 0.5		1.2 ± 1.9		
$5f_{7/2}$-$3d_{5/2}$	4.7 ± 0.8		5.7 ± 2.3		
$4f_{5/2}$-$3d_{3/2}$	19.4 ± 2.8	17.6 ± 3.4	20.8 ± 3.4	18.9 ± 3.5	20.2 ± 5.3
$4f_{7/2}$-$3d_{5/2}$	33.6 ± 4.7	31.7 ± 5.8	35.5 ± 5.6	36.2 ± 6.6	33.3 ± 8.4
8-4	1.1 ± 0.3			2.1 ± 1.0	
7-4	2.0 ± 0.4			3.0 ± 1.0	
6-4	7.1 ± 1.0	5.1 ± 1.1	7.6 ± 1.3	6.5 ± 1.3	6.3 ± 2.1
5-4	42.3 ± 5.9	32.9 ± 10.2	44.1 ± 7.0	58.4 ± 10.5	30.2 ± 10.8
13-5					
12-5					
11-5	1.4 ± 0.3				
10-5	0.8 ± 0.2			1.3 ± 0.7	
9-5	2.4 ± 0.4	1.6 ± 1.2	1.5 ± 0.5	1.3 ± 0.7	
8-5	1.3 ± 0.3		2.2 ± 0.9	0.9 ± 1.1	
α	−0.1	−0.1	−0.1	0.0	−0.1
χ^2_ν	5.8	3.0	1.2	1.4	0.6

TABLE VIII. Intensities of muonic x-ray lines in Sn in several metal halides and pure metals (in %).

Sn	Sn	$SnCl_2$	$SnCl_4$
$2p_{3/2}$-$1s_{1/2}$	60.1 ± 11.9	59.8 ± 9.0	
$2p_{1/2}$-$1s_{1/2}$	34.9 ± 7.1	35.2 ± 6.0	
$3d_{3/2}$-$2p_{1/2}$	23.0 ± 4.0	23.2 ± 3.4	20.5 ± 7.0
$3d_{5/2}$-$2p_{3/2}$	41.8 ± 7.2	43.7 ± 6.1	46.4 ± 13.3
$6f_{5/2}$-$3d_{3/2}$			
$6f_{7/2}$-$3d_{5/2}$			
$5f_{5/2}$-$3d_{3/2}$	2.6 ± 0.7		
$5f_{7/2}$-$3d_{5/2}$	3.8 ± 1.0		
$4f_{5/2}$-$3d_{3/2}$	19.6 ± 3.8	18.9 ± 3.2	
$4f_{7/2}$-$3d_{5/2}$	30.3 ± 6.0	37.6 ± 6.6	
8-4	0.6 ± 0.3	0.7 ± 0.7	
7-4	2.2 ± 0.5	3.1 ± 0.9	
6-4	7.1 ± 1.4	7.1 ± 1.3	
5-4	40.5 ± 7.7	43.3 ± 7.2	
13-5	0.6 ± 0.3	0.4 ± 0.3	
12-5	0.8 ± 0.3	0.8 ± 0.4	
11-5	0.3 ± 0.2	0.2 ± 0.3	
10-5	0.8 ± 0.3	0.3 ± 0.3	
9-5	2.0 ± 0.5	2.0 ± 0.6	
8-5	1.1 ± 0.4		
α	0.1	0.0	
χ^2_ν	4.1	2.2	

TABLE IX. Intensities of muonic x-ray lines in Sb in several metal halides and pure metals (in %).

Sb	Sb	SbF_3	$SbCl_3$	$SbCl_5$
$2p_{3/2}$-$1s_{1/2}$	68.5 ± 16.4	63.8 ± 8.1	65.3 ± 9.0	63.8 ± 11.2
$2p_{1/2}$-$1s_{1/2}$	26.5 ± 7.9	31.2 ± 4.5	29.7 ± 4.7	31.2 ± 6.3
$3d_{3/2}$-$2p_{1/2}$	24.0 ± 5.5	20.5 ± 2.6	22.4 ± 3.1	22.5 ± 4.0
$3d_{5/2}$-$2p_{3/2}$	57.3 ± 13.0	50.2 ± 6.1	53.4 ± 7.1	52.0 ± 8.8
$6f_{5/2}$-$3d_{3/2}$		0.9 ± 0.6		
$6f_{7/2}$-$3d_{5/2}$		1.4 ± 0.9		
$5f_{5/2}$-$3d_{3/2}$	2.9 ± 1.0	2.9 ± 0.9		
$5f_{7/2}$-$3d_{5/2}$	4.7 ± 1.5	4.7 ± 1.2		
$4f_{5/2}$-$3d_{3/2}$	23.3 ± 5.7	19.1 ± 2.9	21.0 ± 3.4	
$4f_{7/2}$-$3d_{5/2}$	40.0 ± 9.7	32.4 ± 4.9	33.2 ± 5.3	
8-4				
7-4	3.3 ± 0.9	3.1 ± 0.8	1.9 ± 0.7	
6-4	8.7 ± 2.1	6.9 ± 1.1	7.2 ± 1.3	
5-4	51.8 ± 12.5	46.2 ± 7.0	46.5 ± 7.3	
13-5				
12-5				
11-5		0.6 ± 0.3		
10-5	0.9 ± 0.4			
9-5	1.9 ± 0.6	1.3 ± 0.3		
8-5	1.2 ± 0.4	1.1 ± 0.5	2.1 ± 0.9	
α	0.1	−0.1	0.0	
χ^2_ν	0.8	2.8	2.5	

TABLE X. Intensities of muonic x-ray lines in I in several metal halides (in %).

I	AlI_3	KI	CdI_2
$2p_{3/2}$-$1s_{1/2}$			49.1 ±7.4
$2p_{1/2}$-$1s_{1/2}$			45.9 ±6.9
$3d_{3/2}$-$2p_{1/2}$	28.8 ±5.1	27.5 ±5.3	27.1 ±3.9
$3d_{5/2}$-$2p_{3/2}$	51.1 ±9.1	52.3 ±9.9	52.8 ±7.4
$6f_{5/2}$-$3d_{3/2}$			
$6f_{7/2}$-$3d_{5/2}$			
$5f_{5/2}$-$3d_{3/2}$	4.2 ±1.4	1.6 ±0.9	
$5f_{7/2}$-$3d_{5/2}$	6.5 ±1.6	4.3 ±1.3	
$4f_{5/2}$-$3d_{3/2}$	26.9 ±4.8	22.6 ±4.3	31.8 ±5.4
$4f_{7/2}$-$3d_{5/2}$	44.7 ±7.9	38.2 ±7.1	50.7 ±8.3
8-4	1.6 ±0.8	1.7 ±0.7	
7-4	4.8 ±1.2	2.6 ±0.8	
6-4	8.5 ±1.6	6.8 ±1.4	11.4 ±2.3
5-4	61.3 ±10.7	50.0 ±9.2	67.1 ±20.0
13-5			
12-5			
11-5			
10-5	1.3 ±0.5	0.7 ±0.5	
9-5	1.7 ±0.5	2.4 ±0.7	
8-5	1.9 ±0.5	2.8 ±1.1	2.6 ±0.9
7-5	8.6 ±1.6	7.2 ±1.5	6.3 ±2.3
α	0.0	0.0	0.1
χ^2_ν	0.5	1.5	0.6

TABLE XI: Pronounced chemical effects in the form of double ratios. Column 1 lists the transitions whose intensity ratios are formed for both compounds listed in the heads of columns 2–4, for the element listed. Columns 2–4 list the corresponding double ratios.

	F: AlF_3/CdF_3	F: AlF_3/SbF_3	Cl: $AlCl_3/CdCl_2$
(3-1)/(2-1)	0.64 ±0.07	0.72 ±0.03	0.54 ±0.08
(4-1)/(2-1)		0.60 ±0.03	

reproduce the experimental data. This means that the concept of these calculations has to be replaced by a new one. It is again too early to say what concept this will be.

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