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Muonic sodium X-ray intensities in different compounds*

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Abstract. Lyman series intensity ratios in muonic sodium have been measured in the ten compounds NaNO_2 , NaNO_3 , Na_2SO_3 , Na_2SO_4 , Na_2SeO_3 , Na_2SeO_4 , Na_2S , Na_2Se , NaCl and NaBr as well as in metallic sodium. No significant differences have been found. From this result conclusions are drawn for the capture and cascade mechanism.

1. Introduction

Recent experiments with muons have shown that both the relative atomic capture probabilities for individual elements in compounds and the relative cascade intensities may be influenced by the chemical structure [1–5]. On the theoretical side, some progress in the understanding of the atomic capture process of muons and other particles has been made [6–9]. However, it is not well known, which parameters (e. g. atomic size, interatomic distance, ionicity of bonds, formal valence states, electron binding energies, 'allotropy') are responsible for the observed effects. On the other hand, even a non-observation of chemical effects can give valuable indications on the atomic capture mechanism and rule out certain hypotheses.

Sodium is one of the elements of the Periodic Table with the smallest electronegativity. Hence, the bonds to other elements or chemical complexes are generally strongly ionic. Only the outermost electron of the sodium atom takes part in bonds so that the formal valence state is always the same. From this point of view, the sodium atom can be considered to behave almost identically in most compounds. By measuring the relative intensities of the Lyman series in muonic sodium in different compounds, we wanted to find out if the initial capture state and hence the subsequent cascade are affected by the bond partners of sodium. The Lyman series is most sensitive to the initial distribution and the electron shell refilling mechanism.

Only a few systematic measurements of muonic X-ray intensities from the same element in different compounds have been performed [1–4]. However, due to the small intensities of the higher members of the Lyman series, definite conclusions regarding chemical effects are not easy to establish.

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In this paper, we present results of Lyman series measurements from muonic sodium up to the $8p \rightarrow 1s$ transitions in the following ten compounds: NaNO_2 , NaNO_3 , Na_2SO_3 , Na_2SO_4 , Na_2SeO_3 , Na_2SeO_4 , Na_2S , Na_2Se , NaCl and NaBr . In order to avoid possible effects due to the transfer mechanism, all compounds were prepared as waterfree samples. For comparison, the Lyman series intensities were also determined in metallic sodium.

2. Experiment and results

The experiment was performed at the muon channel of the SIN accelerator in Villigen. The muonic X-rays were registered in two diodes, a 2.2 cm^3 planar Ge(Li) detector or a 2.4 cm^3 intrinsic Ge detector and a 50 cm^3 coaxial Ge(Li) detector. The in-beam energy resolution of the $2p \rightarrow 1s$ transition in muonic sodium at 250 keV was typically 1.1 keV for the small detectors and 1.7 keV for the large Ge(Li) counter. The detector efficiency calibration at the lower end of

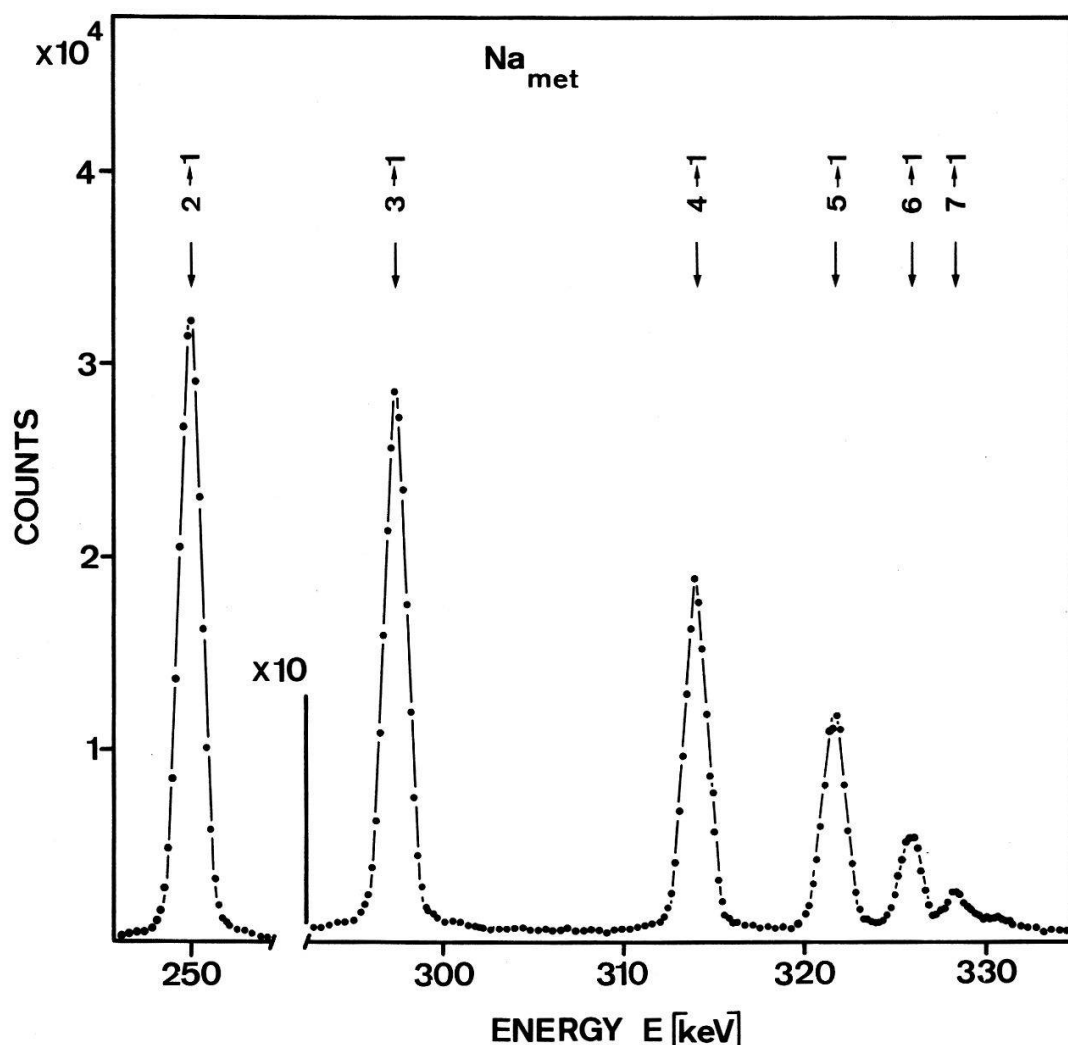


Figure 1
The Lyman series of muonic sodium in the metal, gated by 'prompt' time events (50 cm^3 Ge(Li) detector)

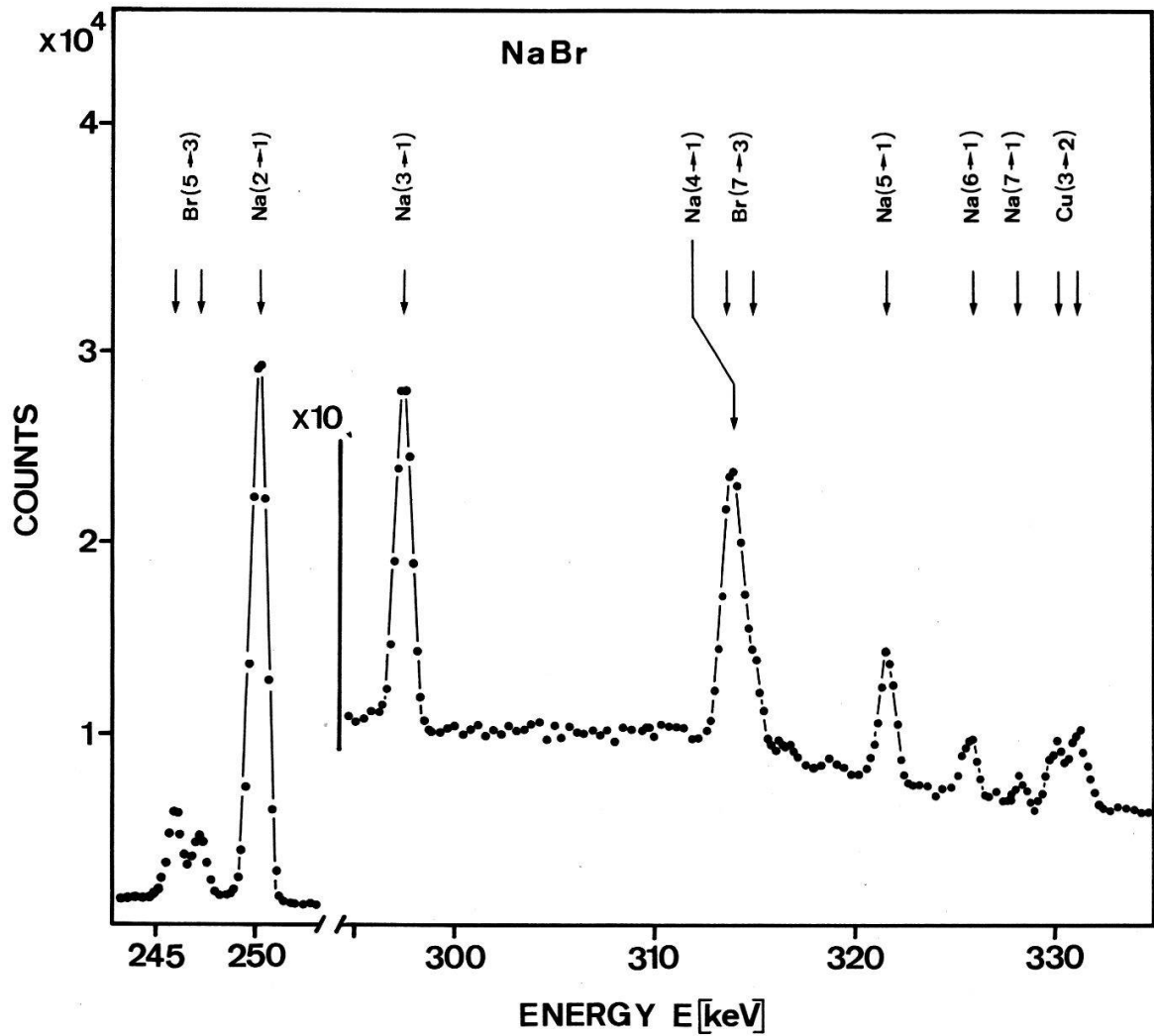


Figure 2

The Lyman series of muonic sodium in NaBr, gated by 'prompt' time events (2.2 cm³ Ge(Li) detector)

the energy scale included the energy dependence of the constant fraction discriminator level in the fast timing branch. Details of the experimental set-up, the data-taking procedure and the analysis of the data may be found in refs. [10] and [11]. Figures 1 and 2 show typical muonic Lyman series spectra in sodium in Na_{met} and NaBr.

Table 1 presents our experimental results for the ten sodium compounds and the sodium metal. All intensities $I(n \rightarrow 1)$ with $n \geq 3$ are normalized to the respective $I(2 \rightarrow 1)$ intensities. The tabulated uncertainties include statistical errors as well as relative errors from the detector efficiency and the self-absorption correction [11] in the respective targets. Our data for NaCl are in agreement with other reported values [1], [4] within the quoted uncertainties.

The main result of our data is the marked similarity of the intensity patterns for all ten compounds and the metallic sodium. This fact is visualized in Fig. 3, where the individual intensity ratios are plotted with respect to the mean value as quoted in Table 1. As a consistency check, both the data taken with the small and the large detector are given in the case of metallic sodium. No systematic deviations from the mean are observed.

Table 1
Lyman series intensity ratios ($\times 10^3$) in muonic sodium

Compound	$(3 \rightarrow 1)/(2 \rightarrow 1)$	$(4 \rightarrow 1)/(2 \rightarrow 1)$	$(5 \rightarrow 1)/(2 \rightarrow 1)$	$(6 \rightarrow 1)/(2 \rightarrow 1)$	$(7 \rightarrow 1)/(2 \rightarrow 1)$	$(8 \rightarrow 1)/(2 \rightarrow 1)$
NaNO ₂	112.0 \pm 5.0	75.5 \pm 4.1	45.1 \pm 3.4	22.8 \pm 2.5	5.0 \pm 1.3	—
NaNO ₃	111.0 \pm 9.0	70.0 \pm 7.0	50.0 \pm 5.0	20.3 \pm 3.0	6.3 \pm 2.0	—
Na ₂ SO ₃	106.0 \pm 3.0	67.0 \pm 3.0	44.0 \pm 2.0	19.0 \pm 2.0	6.0 \pm 1.0	3.0 \pm 1.0
Na ₂ SO ₄	106.0 \pm 3.4	69.2 \pm 2.8	45.8 \pm 2.3	18.8 \pm 1.4	7.3 \pm 1.1	2.2 \pm 1.0
Na ₂ SeO ₃	102.0 \pm 7.0	66.5 \pm 3.0	48.3 \pm 3.0	19.5 \pm 1.0	6.7 \pm 1.0	—
Na ₂ SeO ₄	99.0 \pm 7.0	69.4 \pm 3.0	44.6 \pm 2.0	18.2 \pm 1.5	7.7 \pm 1.0	—
Na ₂ S	113.0 \pm 3.0	75.0 \pm 3.0	48.0 \pm 2.0	21.0 \pm 1.0	8.5 \pm 0.8	1.5 \pm 1.0
Na ₂ Se	107.3 \pm 4.7	71.2 \pm 3.1	46.8 \pm 2.6	21.9 \pm 1.9	6.5 \pm 1.0	—
NaCl	99.4 \pm 7.0	64.8 \pm 4.7	43.0 \pm 3.2	18.3 \pm 1.6	6.9 \pm 0.9	—
NaBr	99.4 \pm 2.6	68.6 \pm 2.6	42.8 \pm 1.6	21.5 \pm 1.3	9.4 \pm 1.4	—
mean value	105.9 \pm 1.8	69.8 \pm 1.3	45.6 \pm 1.0	20.1 \pm 0.6	7.3 \pm 0.4	2.4 \pm 0.6
Na _{met} (a)	109.7 \pm 3.0	72.5 \pm 2.5	46.6 \pm 2.0	19.5 \pm 1.5	8.3 \pm 1.0	2.0 \pm 1.0
Na _{met} (b)	105.5 \pm 3.0	72.5 \pm 2.5	47.9 \pm 2.0	21.3 \pm 1.5	8.3 \pm 1.0	2.4 \pm 1.0

^a measured with 2.2 cm³ Ge(Li) detector

^b measured with 50 cm³ Ge(Li) detector

3. Comparison with cascade calculations

Recent calculations performed under certain restrictive assumptions show [7], [8] that a given distribution over angular momentum states l in a high capture level, e.g. $n = 30$, is only moderately changed in a lower state, e.g. $n = 14$, when the particle cascades down. This means that from measured X-ray intensities one cannot – within the present experimental accuracy – deduce the capture distribution over n -states. However, one can derive trends for the distribution over angular momentum states.

Here, we have tried to reproduce the Lyman series intensities of muonic sodium by a cascade calculation in order to test the sensitivity of the K -series

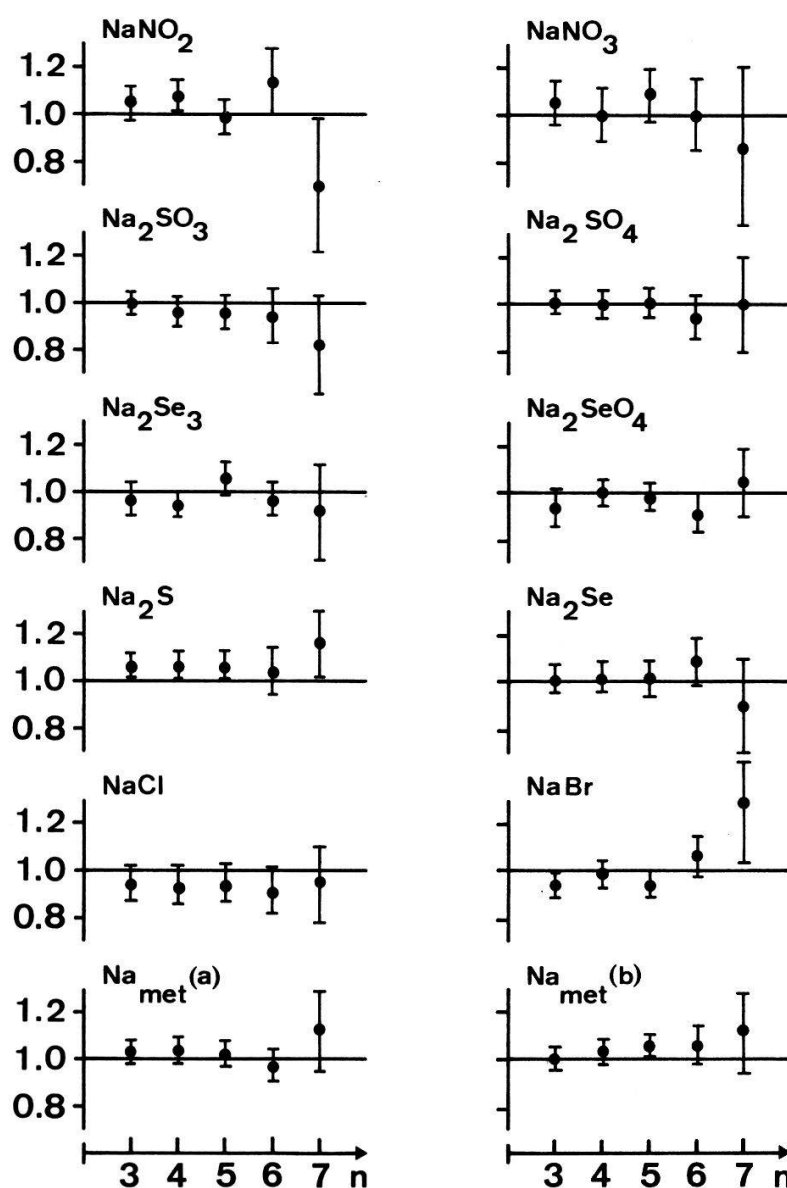


Figure 3

Relative muonic intensity ratios in sodium in the various compounds and in the metal. The points labelled n correspond to the $(n \rightarrow 1)/(2 \rightarrow 1)$ intensity ratio in the compound divided by the corresponding mean values given in Table 1. $\text{Na}_{\text{met}}(\text{a})$ and $\text{Na}_{\text{met}}(\text{b})$ refer to measurements with the 2.2 cm^3 Ge(Li) detector and the 50 cm^3 Ge(Li) detector respectively.

intensities to various assumptions about the angular momentum distributions in an assumed initial capture level $n = 14$. Such a test may be performed using a recently published computer code [12]. Since a modified older cascade computer code still in general use [13] yielded similar results on the muonic cascade in thallium [12], needs much less computer time and has less free or adjustable parameters, we have calculated the Lyman series intensities of muonic sodium using this older code.

The following assumptions about initial angular momentum distributions have been made:

- I. Homogeneous distribution: $\text{pop}(l) = \text{const.}$
- II. Statistical distribution: $\text{pop}(l) \propto (2l + 1)$
- III. Modified statistical distribution: $\text{pop}(l) \propto (2l + 1) \cdot \exp(\alpha l)$
- IV. Linear combinations of distributions I. and III.

Figure 4 shows the comparison between such cascade calculations and the experimental sodium data. The bars represent the total spread of the measured relative intensities (not the mean values). With conventional angular momentum distributions, the observed experimental values cannot be reproduced (curves *a* to *e*). A linear combination (curve *f*, two adjustable parameters) gives already a better, but still unsatisfactory agreement with the experimental data. Only 'unconventional' distributions like a 10% population of the $l = 0$ state plus modified statistical distributions (curves *g* and *h*, three adjustable parameters) reproduce the experimental values well. However, these parameters are not unique (five experimental values reproduced with three parameters) and one needs further experimental transition intensities (*L*-series) in order to draw conclusions about the initial angular momentum distribution.

4. Discussion

The non-observation of significant differences in the muonic Lyman series intensities in sodium in the various compounds and in the sodium metal rules out some hypotheses regarding the atomic capture mechanism of muons and their subsequent cascade.

For example, one could imagine that simply the bond to an element of another kind changes the initial capture distribution or the subsequent cascade. Differences in the Lyman series have been found, e.g., in titanium in Ti_{met} and TiO_2 [14], [15] and in nitrogen in BN compared to NaNO_2 and NaNO_3 [3]. Our result disproves such an hypothesis.

The valence electron in sodium in the metallic form belongs to the electron gas, while in a compound it is shared with the bond partner(s). In all our investigated materials including Na_{met} , sodium can practically be considered as a sodium ion. From our results, one may thus deduce that the electron gas does not actively take part in the capture mechanism.

The way the captured muon cascades down is governed by the presence of the electrons in the atom. In the early stage of the cascade, Auger electron transitions prevail. If the electron hole refilling is slower than the cascade transitions, radiative transitions begin to predominate over Auger electron ejection. The population of circular orbits then increases and the intensities of the

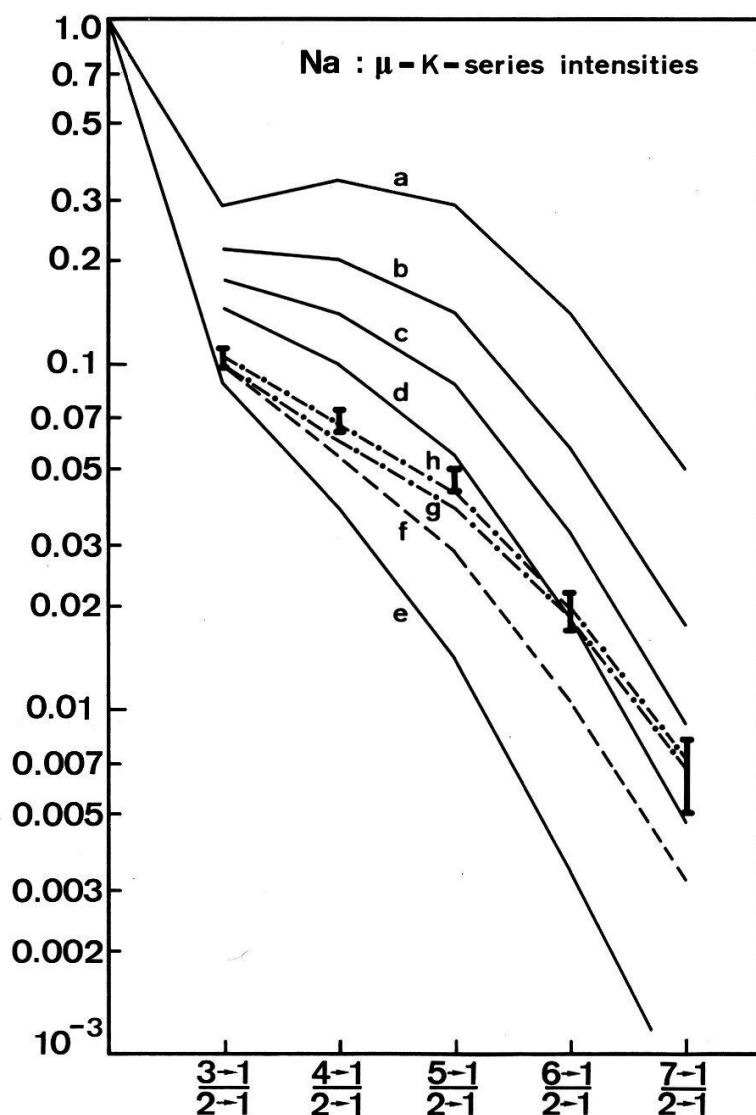


Figure 4

Comparison of experimental muonic intensity ratios in sodium. The bars represent the total spread of the measured relative intensities. The different lines correspond to the following assumptions about the initial angular momentum distribution in the $n = 14$ level:

- a) homogeneous distribution: $\text{pop}(l) = \text{const.}$
- b) statistical distribution: $\text{pop}(l) \propto (2l+1)$
- c) modified statistical distribution: $\text{pop}(l) \propto (2l+1) \exp(\alpha l)$ with $\alpha = 0.10$
- d) $\text{pop}(l) \propto (2l+1) \exp(\alpha l)$ with $\alpha = 0.20$
- e) $\text{pop}(l) \propto (2l+1) \exp(\alpha l)$ with $\alpha = 0.50$
- f) $\text{pop}(l) \propto 0.1 + 0.9(2l+1) \exp(\alpha l)$ with $\alpha = 0.50$
- g) $\text{pop}(l=0) = 0.1$, $\text{pop}(l>0) \propto (2l+1) \exp(\alpha l)$ with $\alpha = 0.50$
- h) $\text{pop}(l=0) = 0.1$, $\text{pop}(l>0) \propto (2l+1) \exp(\alpha l)$ with $\alpha = 0.45$

higher members of the Lyman series are correspondingly decreased. In sodium metal, the electron hole refilling mechanism is faster than in the insulators. However, no difference has been found in the muonic X-ray intensities although the investigated materials range from good conductors to good insulators. The differences in the electron refilling times seem therefore not to give measurable changes in the muonic Lyman series intensities.

Correlations have been found between the X-ray intensity structure of

captured kaons and the atomic radius [16–18]. However, in compounds and allotropic modifications of an element [11], the atomic radius is somewhat difficult to define in a unique way, e.g. the carbon radius, where one has single, double or triple bonds. The interatomic distance seems therefore to be a more reliable correlation parameter. In the investigated compounds however, the interatomic distances between sodium and the nearest partners are different. Typical values are: 2.83 Å for Na—S in Na₂S and 2.45 Å for Na—Cl in NaCl or 2.47 Å for Na—O in NaNO₂ and 2.31 Å for Na—O in Na₂SO₄. Hence, the interatomic distances vary by more than 20% without a measurable change in the muonic X-ray intensity pattern.

One could imagine that the energy spectrum of slow muons in matter were different due to different relative concentrations of various elements in the target. In that case, one would expect differences in the initial angular momentum distribution and perhaps also in the capture probabilities [19]. However, a recent calculation [20] has shown that a concentration effect on capture ratios in binary mixtures is negligibly small as long as the energy losses in meson-atom collisions are not much smaller than the centrifugal barrier height.

From the point of view of the electronic structure, the sodium atoms in all investigated compounds as well as in the metal can be considered as sodium ions Na⁺ with a constant ionic radius of 0.97 Å. Indeed, according to Pauling [21], the sodium bonds in all studied targets are strongly ionic (more than 40% ionic character), and the metallic sodium can be regarded as sodium ions embedded in an electron gas. Such a similarity in the behaviour of sodium in the investigated targets may well explain the absence of so-called chemical effects. Observed chemical effects in other elements (cf. e.g. ref. [22] and references cited therein) must therefore be correlated to the chemical structure of a compound, i.e. to the valence electron distribution among bond partners. Such an interpretation also fits into the general framework of a recently developed model for the atomic capture process of negative exotic particles [22], [23].

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