Electronic structure and muonic x-ray intensities in isoelectronic series of neon and argon

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We report on muonic Lyman-series x-ray intensities measured in gaseous argon and in K^+, Cl^-, Na^+, and F^- in the compounds KF and NaCl. The analysis of their structures, including neon, shows that both the spatial symmetry and the binding energy of the electrons of outer shells determine the angular momentum distribution of captured muons. The conclusions are in agreement with the modified model of "large mesic molecules."

INTRODUCTION

Most experimental results on muonic atom formation reported so far concern capture ratios in compounds and mixtures. These ratios were used to construct and to test models for the capture mechanism. The muonic x-ray intensities measured to determine capture ratios were also analyzed. Systematic comparisons have shown correlations to chemical parameters.[1-10] However, Naumann et al.[10] observed that muonic x-ray intensity patterns are insensitive to the charge of the ionic core.

Muonic x-ray intensity patterns give information on the muonic atom formation mechanism. In order to gain greater insight into the angular momentum distribution of captured muons, the measured x-ray intensities have to be fitted using cascade calculations. Direct comparisons of muonic x-ray intensities may disagree, but similarities are discovered through cascade calculations. For example, the muonic intensity ratio $I(3\text{-}1)/I(2\text{-}1)$ in the ionic compound K^+F^- is about two times larger in fluorine than in potassium, although the electronic distributions are very similar. The fit of the measured x-ray intensities revealed indeed a statistical angular momentum distribution at $n = 14$ in both cases.

The aim of the present work was to determine the "initial" angular momentum distribution of captured muons in the noble gases argon and neon. The electrons in noble gases are distributed in closed subshells which have central symmetry with regard to the nucleus. There is no bond between atoms, such that chemical effects do not influence the muon capture at all. The distributions are then compared to the distributions fitted in isoelectronic ions of Cl^- and K^+ on one side and F^- and Na^+ on the other side.

EXPERIMENTAL RESULTS

Our measurements were performed at the superconducting muon channel of the Swiss Institute for Nuclear Research (SIN) in Villigen, Switzerland. The experimental setup, the data acquisition system, and the data analysis were comparable to earlier experiments.[7,11] The muonic x-rays were registered either by a 50- or 66-cm^3 Ge detector.

Muonic argon x-ray intensities have been measured in pure gaseous argon at three different pressures. At 22 and 170 bars an aluminum pressure vessel was employed which was originally constructed for measurements of muonic isotope shifts in noble gases[12] using the muon transfer from hydrogen. The measurement at 1.5 bar was performed with a stainless-steel vessel which will be described elsewhere.

The muonic x-ray intensities in K^+, F^-, Na^+, and Cl^- have been measured in the compounds KF and NaCl. The target materials were contained in a flat cylindrical beryllium box. The targets with thicknesses of 1 g/cm^2 were placed at 45° relative to the muon beam and viewed under 45° by the Ge detector. The efficiency calibration of the detectors was performed using ^{75}Se, ^{133}Ba, ^{152}Eu, and ^{166}Ho gamma-ray sources.[13]

Our measured intensity patterns of the Lyman series in argon (Table I) confirm the target density effects on the muonic cascade already observed by Knight et al.[14] and in neon by Ehrhart et al.[15] (Table II). These effects are principally characterized by an enhancement of the muonic n-1 intensities with regard to the (2-1) line at higher pressures, where electron transfer rates by collisions are larger. With the exception of the argon (6-1) line, all other K-series intensities up to (7-1) could be precisely determined. Regarding the argon (6-1) transition, the 846.77-keV gamma line resulting from the nuclear excitation of ^{56}Fe could not be sufficiently resolved. The given intensity for this line (Table I) is therefore only an estimate and has not been taken into account in the fit of the Lyman-series intensities.

The muonic Na^+ intensities (Table III) from a new measurement[16] in NaCl agree with those published earlier.[2,4] The same is true for the chlorine x rays.[4] The muonic potassium x-ray intensities in KF agree with those measured in KCl (Refs. 4 and 17) (Table IV).

TABLE I. Intensities of the Lyman series in argon and reproduction of the muonic cascade with a statistical initial distribution at $n = 14$.

	Expt. 1.5 bar	Calc. $P_L^e = 2\%$	Expt. 22 bar	Calc. $P_L^e = 3.5\%$	Expt. 170 bar	Calc. $P_L^e = 15\%$
$2p$-$1s$	0.914(26)	0.909	0.905(25)	0.899	0.875(24)	0.870
$3p$-$1s$	0.0400(22)	0.0395	0.0410(14)	0.0427	0.0537(16)	0.0509
$4p$-$1s$	0.0097(11)	0.0112	0.0116(8)	0.0128	0.0187(8)	0.0185
$5p$-$1s$	0.0083(8)	0.0066	0.0095(6)	0.0085	0.0154(6)	0.0161
$6p$-$1s$	(0.0080)	0.0065	(0.0094)	0.0086	(0.0133)	0.0155
$7p$-$1s$	0.0077(8)	0.0075	0.0093(6)	0.0090	0.0111(5)	0.0119
χ^2		1.31		1.36		1.42

TABLE II. Intensities of the Lyman series in neon by Ehrhart *et al.* (Ref. 15) and reproduction of the muonic cascade with a statistical initial distribution at $n = 14$.

	Expt. 0.4 bar	Calc. $P_L^e = 0.30\%$	Expt. 5.9 bar	Calc. $P_L^e = 0.50\%$	Expt. 35.2 bar	Calc. $P_L^v = 0.55\%$
$2p$-$1s$	0.9183(74)	0.9110	0.9011(89)	0.9006	0.9009(41)	0.8982
$3p$-$1s$	0.0383(44)	0.0387	0.0397(60)	0.0426	0.0425(33)	0.0433
$4p$-$1s$	0.0105(30)	0.0114	0.0130(17)	0.0134	0.0134(10)	0.0139
$5p$-$1s$	0.0056(23)	0.0063	0.0097(49)	0.0083	0.0074(14)	0.0088
$6p$-$1s$	0.0061(22)	0.0057	0.0097(44)	0.0077	0.0122(13)	0.0081
χ^2		0.24		0.12		2.34

TABLE III. Intensities of the Lyman series in NaCl and muonic cascade with a statistical initial distribution at $n = 14$.

	Sodium					Chlorine		
	Piller 85[a] NaCl	Daniel 81[b] NaCl	Calc. $P_L^e = 20\%$	Calc. $P_L^e = 30\%$	Calc. $P_L^e = 40\%$	Piller 85[a] NaCl	Daniel 81[b] NaCl	Calc. $P_L^e = 30\%$
$2p$-$1s$	0.803(17)	0.814(7)	0.790	0.774	0.762	0.819(19)	0.830(27)	0.835
$3p$-$1s$	0.0824(22)	0.081(4)	0.0779	0.0863	0.0919	0.0661(22)	0.0670(20)	0.0636
$4p$-$1s$	0.0527(14)	0.052(3)	0.0556	0.0626	0.0677	0.027 33(83)	0.0272(10)	0.026 90
$5p$-$1s$	0.034 40(98)	0.034(2)	0.0407	0.0438	0.0461	0.026 50(85)	0.0255(10)	0.025 36
$6p$-$1s$	0.015 58(42)	0.015(1)	0.0206	0.0204	0.0205	0.020 56(69)	0.020 59(9)	0.021 40
$7p$-$1s$	0.005 78(24)	0.0034(6)	0.0086	0.0078	0.0074	0.013 11(47)		0.013 43
χ^2	Piller 85[a]		48.7	55.5	75.8			1.00
	Daniel 81[b]		21.9	25.7	33.6			0.81

[a]Reference 16.
[b]Reference 4.

TABLE IV. Intensities of the Lyman series in KF (KCl) and muonic cascade with a statistical initial distribution at $n = 14$.

	Potassium				Fluorine	
	This work KF	Daniel 81[a] KCl	Naumann 80[b] KCl	Calc. $P_L^e = 30\%$	This work KF	Calc. $P_L^e = 45\%$
$2p$-$1s$	0.829(25)	0.833(24)	0.845(23)	0.834	0.731(29)	0.730
$3p$-$1s$	0.0638(21)	0.062(4)	0.068(3)	0.0662	0.117(5)	0.121
$4p$-$1s$	0.0242(10)	0.033(3)	0.026(2)	0.0243	0.0876(36)	0.0860
$5p$-$1s$	0.0227(9)	0.020(3)	0.024(1)	0.0216	0.0451(20)	0.0425
$6p$-$1s$	0.0210(9)	0.028(4)	0.018(1)	0.0199	0.0151(8)	0.0139
$7p$-$1s$	0.0133(6)	0.013(3)	0.014(1)	0.0142	0.0034(7)	0.0043
χ^2	This work			1.01		1.07
	Daniel 81[a]			2.43		
	Naumann 80[b]			1.61		

[a]Reference 4.
[b]Reference 17.

CASCADE CALCULATIONS

The muonic level n, where a cascade calculation is started, is to a certain extent arbitrary. Starting the cascade calculation with a given angular momentum distribution at $n = 14$, 16, or 18, the calculated x-ray intensities are not significantly different. Thus, we have chosen as initial level $n = 14$ and assumed there a statistical initial angular momentum distribution to try to reproduce the muonic x-ray intensities of the Lyman series measured in argon and neon, and in the Na^+, Cl^-, K^+, and F^- ions. In addition, it has been tested whether a simultaneous fit of the intensities of the Lyman and Balmer series gives the same result as a fit of the Lyman series alone. Indeed, the inclusion of the Balmer series intensities in chlorine[16] and chromium[18] does not significantly change the initial angular momentum distribution of the captured muons. Since the pattern of the Lyman-series intensities is more sensitive to differences in the initial angular momentum distributions of the muons than the Balmer series, such a result is expected.

We employed the cascade program MUON00 written by Akylas and Vogel.[19] The binding energies of the electrons of the Z-1 elements have been taken from the tables of Sevier,[20] the effective nuclear charges for the K, L, and M shells from Clementi and Raimondi,[21] and the K-shell refilling widths Γ_0 for the neutral atoms from McGuire.[22]

During the muon cascade, part of the electronic L and M shells are emptied and the K-shell refilling width Γ_K becomes consequently smaller. In the cascade calculation, Γ_K was correlated linearly to the electronic L-shell population using the relation

$$\Gamma_K = \Gamma_0 P_L^e , \tag{1}$$

where P_L^e is the L-shell population at the beginning of the cascade calculation relative to its population in the neutral atom. This correlation is an acceptable approximation and allows to reduce the number of adjustable parameters in the fit of the measured Lyman-series intensities.

As already mentioned (see Tables I and II), the muonic x-ray intensity patterns in gases are pressure dependent. But on the other hand, we expect that the initial angular momentum distribution of the captured muons is independent of the pressure, and that the differences in the intensity patterns are due to the refilling rate of the electron shells, which depends on the collision rate.[23] In fitting the muonic intensities in the gases, we assumed that the L and M electrons ejected during the muon cascade are not replaced. The L-shell population, P_L^e, is then a global measure of the mean ionic charge of the element and of the collision rate.

The muonic x-ray intensities of the K^+, Cl^-, Na^+, and F^- ions were measured in compounds of crystalline structure, where the electron refilling is fast. Hence, we assumed that the populations of the M and L shells remain constant during the cascade.

In a first series of fits of the measured Lyman-series intensities in argon, the electronic L-shell population, P_L^e, was varied and the parameter α of the modified statistical angular momentum distribution
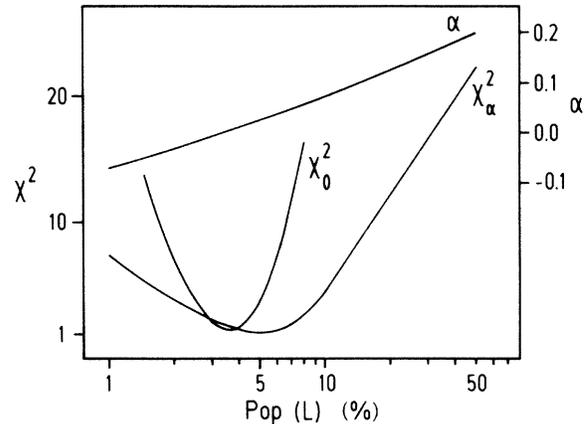
FIG. 1. Fitted values of the parameter α of the modified statistical angular momentum distribution and corresponding X_α^2 values for the muonic Lyman-series intensities in argon at 22 bars in dependence of the electronic L-shell population, $Pop(L) = P_L^e$. For comparison, X_0^2 values for purely statistical distribution ($\alpha = 0$) are also drawn.

$$P(l) \propto (2l + 1) \exp(\alpha l) , \tag{2}$$

adjusted to obtain minimal X^2 values. Figure 1 shows the normalized X^2 values and the fitted values of the parameters α as a function of P_L^e for argon at 22 bars. The X^2 values obtained assuming a purely statistical distribution ($\alpha = 0$) are drawn too. The comparison of the two X^2 curves reveals that the experimental intensities can be reproduced as well with a purely statistical distribution. Similar series of fits have been performed for other elements. The electronic L-shell population, P_L^e, was then the only adjustable parameter in the fit of the measured intensities. The result obtained for argon at 22 bars seems in contradiction with the analysis of Knight et al.[14] performed on muonic data measured in argon at 20 bars. One observes, however, that their $Ki/K\alpha$ intensities are systematically lower than ours and that their analysis pursued another aim.

In Tables I and II, we compare the experimental muonic x-ray intensities of the Lyman series for argon and neon[15] measured at different pressures with intensities calculated by using the cascade program MUON00. There, a statistical initial angular momentum distribution in the muonic $n = 14$ level is assumed and only the electronic L-shell population parameter, P_L^e, is adjusted as discussed above. The adjustment of this one parameter suffices to reproduce well the measured intensities.

Tables III and IV show a comparison of measured muonic x-ray intensities of the Lyman series in the Na^+, Cl^-, K^+, and F^- ions with calculated ones by adjusting again only the P_L^e parameter. With the exception of the muonic Na^+ intensities, all others are well reproduced under the assumption of a statistical initial angular momentum distribution for the captured muons.

DISCUSSION

In gases, the parameter P_L^e decreases with decreasing pressure. Such a behavior is expected. It corresponds to

an increase of the mean ionization of the muonic atom with decreasing collision rate. In neon, the value of this parameter corresponds to an almost fully ionized muonic atom.[23] In argon, which has more electrons in its neutral state, this parameter is, at comparable pressures, nearly one order of magnitude greater.

In solids, specifically for the Cl^-, K^+, and F^- ions, the populations of the L shell at beginning of calculations are mutually comparable, but much larger than in gases. The electronic shells depopulated through muonic Auger transitions are there refilled by the nearest bond partners.

The Cl^- and K^+ ions have the same number of electrons and the same electronic configurations as the neutral argon atom. The binding energies of their electrons and their ionic radii are however different. The difference in the ionic radii[24] between Cl^- (1.81 Å) and K^+ (1.33 Å) is of the order of 30%. Nevertheless, the measured muonic intensities can be reproduced by assuming the same initial angular momentum distribution in the muonic $n = 14$ level. Consequently, the size of the ionic radius does not influence the initial angular momentum distribution of captured muons. In ions with closed-shell electron configurations, the relative insensitiveness of the capture ratios to the ionic radius has already been noticed.[10]

It is remarkable that the initial angular momentum distribution is statistical both in Cl^- and K^+ as well as in argon. Indeed, the muonic x-ray intensities have been measured in the compounds KCl, KF, and NaCl, which are so-called ionic crystals, but where the valence electron transfer is, nevertheless, not complete. The ionicity of the bond, which can be considered as a measure of the electron transfer is, following Phillips,[25] rather high, i.e., 0.953, 0.955, and 0.935, respectively. According to Pauling,[26] the respective values are 0.70, 0.92, and 0.67 for the three compounds. These variations of ionicity for such medium Z elements in ionic crystals do not notably affect the angular momentum distribution of the captured muons. If one assumes that the statistical initial angular momentum distribution reflects the central symmetry of the electrons through which the muons are captured, we have to conclude that the spatial electron distribution in Cl^- and K^+ in the compounds is sufficiently similar to that in argon.

In neon, one observes the same statistical initial angular momentum distribution as in argon, although there are no M-shell electrons, and the neon radius is smaller by 30%. The muonic x-ray intensities of the F^- ion can also be reproduced by a cascade calculation with a statistical initial distribution. The same should be true for the muonic Na^+ ion. Indeed, if in the isoelectronic, centrally symmetric series Cl^-, Ar, and K^+ the measured muonic x-ray intensities can be reproduced with a statistical initial distribution, the lower Z series F^-, Ne, and Na^+, which has the same electronic characteristics, should show the same behavior. But the muonic x-ray intensities of Na^+ cannot be fitted with such a statistical distribution (Table III). This has already been shown by different authors.[1,2,27] In order to fit the muonic x-ray intensity structure in the Na^+ ion measured in various compounds, one needs the modified statistical initial angular

momentum distribution. The P_L^e parameter, however, is still comparable to that of Cl^- and K^+. The value of $\alpha = +0.054$ gives an initial distribution which is significantly different from a statistical one, and which favors higher l states.

The reason for such a nonstatistical initial distribution in Na^+ cannot be found in the chemical properties of sodium or the chemical bond of NaCl, otherwise one should observe nonstatistical initial distributions both in K^+ and in Cl^-.

In the modified version of the model of "large mesic molecules,"[28,29] one assumes that only outer, loosely bound electrons are responsible for muon capture. For the sake of simplicity, a sharp cut is introduced in the electron binding energies, such that all electrons with binding energies less than 60 eV participate with equal weight in the muon capture, and those with higher binding energies do not participate at all. Although such a sharp step function seems unphysical, the experimental muon capture ratios in low-Z elements are satisfactorily reproduced.

Following the model, muons would be captured in Cl^-, Ar, and K^+ exclusively through the complete electronic $3s$ and $3p$ subshells, in F^- and Ne exclusively through the complete $2s$ and $2p$ subshells, whereas in Na^+ the muon capture would proceed only through the complete $2p$ subshell since the binding energy of its $2s$ electrons is 63.4 eV.[20] If the muonic x-ray intensities reflect characteristics of the electrons through which muon capture takes place,[28] one might argue that the capture through complete $2s$ and $2p$ subshells or complete $3s$ and $3p$ subshells produces a statistical initial angular momentum distribution for the captured muon. The initial angular momentum distribution of the muons captured in Na^+ must then reflect the capture through $2p$ electrons alone. The corresponding distribution populates higher l states than the statistical distribution.

If one assumes that the statistical distribution $P_{st}(l)$ is a linear combination of the distribution $P_s(l)$ due to capture through s electrons and the distribution $P_p(l)$ due to capture through p electrons in the outer shell, one can write

$$P_{st}(l) = aP_s(l) + bP_p(l) \tag{3}$$

with

$$\sum_l P_{st}(l) = \sum_l P_s(l) = \sum_l P_p(l) = 1 \tag{4}$$

and

$$a + b = 1 , \tag{5}$$

where a is the probability of muon capture by the two s electrons and b by the six p electrons.

Since the modified model of large mesic molecules predicts rather well the capture probabilities in low-Z atoms, the assumption made there can be used to determine the coefficients a and b. With the hypothesis that only electrons with binding energies less than 60 eV capture muons, one obtains the same values for Cl^-, Ar, K^+, F^-, and Ne, namely $a = 0.25$ and $b = 0.75$. As stated before,
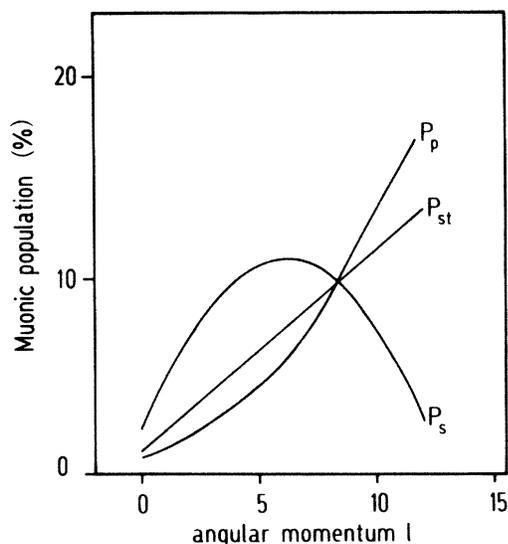
FIG. 2. Contribution to muonic angular distribution by each subshell s and p.

the muonic capture in these ions or elements proceeds through filled $2s, 2p$ or $3s, 3p$ subshells. For Na^+, one has $a = 0$ and $b = 1$. A smooth boundary instead of a sharp cut in the binding energies of the electrons responsible for muon capture seems, however, physically more realistic. The analysis of muon capture ratios in $NaClO_x$,[16] Na_2SO_x, and Na_2SeO_x compounds,[30] where the sodium bond is ionic to nearly 100%, suggests that the participation of the $2s$ electrons of sodium remains weak, such that $a \ll b \cong 1$. Thus, taking for $P_p(l)$ the modified statistical distribution determined in Na^+, an approximate $P_s(l)$ distribution can be calculated. Figure 2 shows that $P_s(l)$ favors low-l states.

The model by von Egidy, Jakubassa-Amundsen, and Hartmann[31] assumes also that only electrons with binding energy lower than a given limit (~ 100 eV) take part in muon capture, their weight decreasing with increasing binding energy. However, this model, which gives good predictions for capture ratios in compounds with $Z > 5$, does not allow to explain both the similarities in the muonic x-ray intensity structure between Ne and Ar, and the difference between Na^+ and the other elements.

CONCLUSION

The analysis of the muonic x-ray intensity patterns in the isoelectronic series Cl^-, Ar, and K^+ suggests that the

central symmetry distribution of the outer electronic shell, where muons are captured, is responsible for the observed statistical initial angular momentum distribution in a muonic $n = 14$ level. In the comparable isoelectronic series F^-, Ne, and Na^+, only sodium does not show such a statistical initial distribution. The explanation for this exception cannot be found in chemical properties of sodium nor in the chemical bond of NaCl, where the muonic x-rays were measured.

Models for the Coulomb capture of negative muons assume that only those electrons contribute to the capture, which have binding energies less than a given limit.[28,31] Such models help in the interpretation of similarities and differences in muonic x-ray intensity patterns. They suggest that the observed statistical initial angular momentum distribution in a given muonic level $n = 14$ might be related to the muon capture through full s and p subshells. The exception of Na^+ can be explained by assuming a rather sharp cut around 60 eV in the binding energies of the electrons contributing to capture.

Following the modified model of large mesic molecules,[28,29] the initial angular momentum distribution observed in Na^+ corresponds to a distribution due to muon capture via the full p subshell alone. Assuming that the statistical distribution observed in the other elements is a linear combination of the distribution via the p subshell and the distribution via the s subshell, one can determine the angular momentum distribution of muons captured within the full s subshells (see Fig. 2). Hence, if the analysis of muonic x-ray intensity patterns is combined with basic assumptions of models for the muonic Coulomb capture, further progress in the understanding of the formation mechanism of exotic atoms can be made.

The capture of negative muons in the Coulomb field of nuclei via atomic electrons is only part of our investigations. Muonic x-ray intensities have also been measured in cubic and hexagonal structures of diamond and boron nitride,[7,9] and in phosphorus and selenium modifications,[32] where the structure of bonding electrons is important. A comparable analysis about muon capture through bonding electrons will be submitted later for publication.

ACKNOWLEDGMENTS

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