Empirical Intensity Correlations in Muonic X-Ray Spectra of Oxides

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The Lyman intensities of muonic x rays from 35 oxides have been measured with Ge detectors. The dependence of the various quantities on each other and on target data is investigated with correlation theory. Many correlations are established.

Although the Coulomb capture of muons and the subsequent x-ray cascade have been found to be of great interest in the last years, the detailed dependence of capture ratios and line intensities on the atomic number Z and the chemical form remained unexplained.[1] The per-atom capture ratios $A(Z_1/Z_2)$ of oxides ($Z_2=8$) show a periodic variation with Z_1 of the oxidized element.[1-3] Similar behavior in muonic, pionic, and kaonic x-ray yields has been observed and discussed.[4-7] It is the aim of this Letter to present new experimental data with muons on oxides and to examine to what extent the various empirical quantities are correlated to each other and to target data. In this examination, correlation theory will be applied.

The experiment was performed at Schweizerisches Institut für Nuklearforschung, Villigen, Switzerland, and consisted in measuring muonic Lyman series of 35 oxides from $Z_1=11$ to $Z_1=52$ with Ge detectors, as described previously.[8] Figure 1 shows a typical spectrum. The line intensities were evaluated and corrected for self-absorption and detector efficiency averaged over the target geometry. The ratios $A(Z_1/Z_2)$ were obtained by summing up the Lyman-series intensities.

Part of the experimental data is shown in Fig. 2. I_ν [$I_{\rm rest}$] means the fraction of the total Lyman intensity of the element falling in the line $\nu \to 1$ [$\Sigma(\nu\to 1)$ for $\nu>3$ (oxygen) or $\nu>4$ (oxidized element), respectively]. The superscripts "met" and "ox" refer to oxidized element and oxygen, respectively. For comparison the atomic radii R_0 for condensed matter[9] are also displayed. A full account of the work giving details of the experiment and the individual experimental numbers will later be given elsewhere.[10]

As can be seen from Fig. 2 there are correlations between experimental quantities. For example, $I_{\rm rest}{}^{\rm met}$ is, on the average, large at the same Z_1 at which $A(Z_1/Z_2)$ is also large. In order to obtain numbers for the degree of correlation we apply mathematical correlation theory

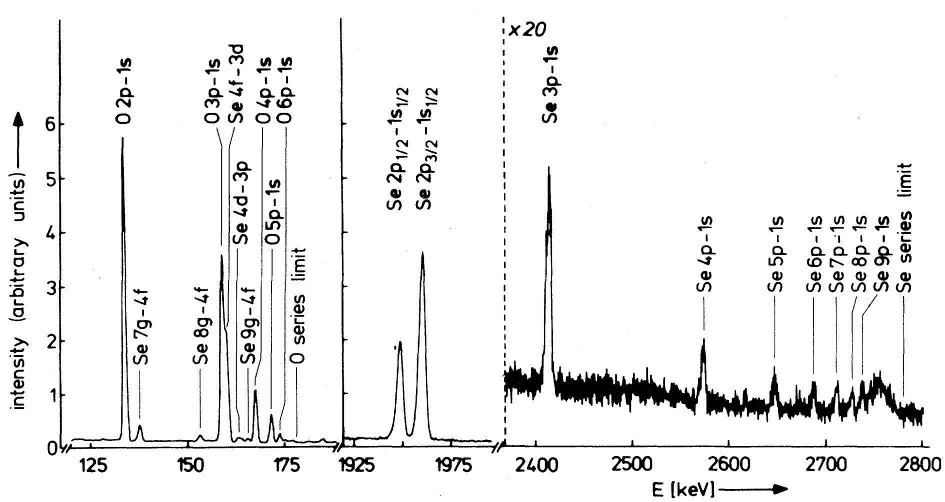

FIG. 1. Muonic x-ray spectrum of SeO_2. O, Lyman region: high-resolution small diode; Se, Lyman region: high-efficiency large diode.

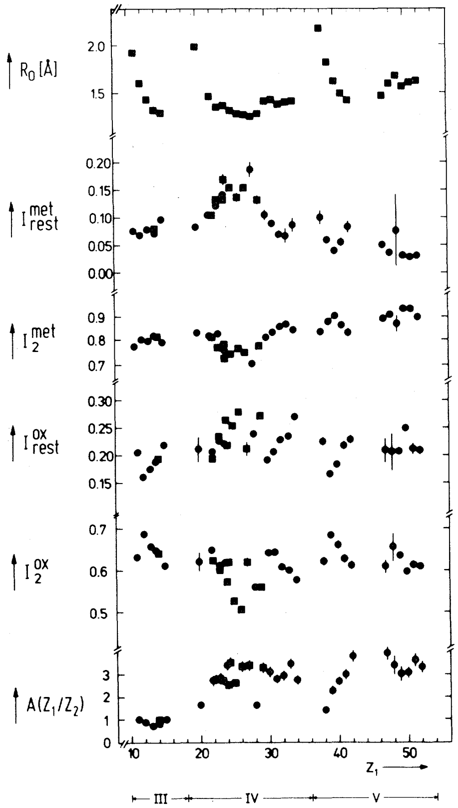

FIG. 2. Capture ratios $A(Z_1/Z_2)$, selected intensities I, and atomic radii R_0 vs atomic number Z_1 of oxidized element. Circles: highest-oxidation state; squares: lower-oxidation states. The periods of the Periodic Table are indicated. For a full explanation see text.

which, to the best of our knowledge, has never been applied before in this or a related field of physics. Note that the correlated values are *not* from quantities measured in coincidence.

Following van der Waerden,[11] we form the empirical correlation coefficient

$$r_{xy} = \frac{\sum_{i=1}^{n}[(x_i - \langle x\rangle)(y_i - \langle y\rangle)]}{[\sum_{i=1}^{n}(x_i - \langle x\rangle)^2]^{1/2}[\sum_{i=1}^{n}(y_i - \langle y\rangle)^2]^{1/2}}, \quad (1)$$

where (x_i, y_i) are data pairs at the same value of Z_1 for two different quantities x and y, such as $x = I_{rest}^{met}$ and $y = A(Z_1/Z_2)$; n is the number of investigated oxides ($n = 35$ in this experiment); and $\langle x\rangle$ and $\langle y\rangle$ denote the mean of all x_i and y_i, respectively. In order to eliminate a supposed dependence on a third quantity z with individual values z_1, we form the partial empirical correlation coefficient

$$r_{xy|z} = \frac{r_{xy} - r_{xz}r_{yz}}{(1 - r_{xz}^2)^{1/2}(1 - r_{yz}^2)^{1/2}}. \quad (2)$$

In our analysis z will be Z_1 or R_0. The reason for eliminating a Z_1 dependence is that many atomic properties depend on Z, and hence correlations between two other quantities may arise from their common Z dependence only. The reason for eliminating an R_0 dependence is that R_0 is expected to have a large geometrical effect which may mask effects with more physical meaning.

TABLE I. Empirical correlation coefficients. Above principal diagonal: r_{xy}. Below principal diagonal: $r_{xy|z}$ with $z = Z_1$.

	Z_1	R_0	B	I_2^{ox}	I_3^{ox}	I_{rest}^{ox}	I_2^{met}	I_3^{met}	I_4^{met}	I_{rest}^{met}	$A(Z_1/Z_2)$
Z_1		.22	-.31	-.09	.07	.09	.73	-.70	-.81	-.51	.66
R_0	—		.60	.39	-.30	-.35	.43	.25	-.09	-.46	-.28
B	—	.72		.43	-.26	-.44	-.18	.17	.31	.10	-.38
I_2^{ox}	—	.42	.43		-.74	-.92	.33	-.05	.23	-.48	-.38
I_3^{ox}	—	-.32	-.25	-.74		.43	-.27	.10	-.05	.33	.18
I_{rest}^{ox}	—	-.38	-.43	-.92	.43		-.29	.01	-.29	.46	.41
I_2^{met}	—	.40	.08	.58	-.47	-.52		-.72	-.57	-.93	.26
I_3^{met}	—	-.14	-.08	-.16	.21	.10	-.42		.68	.45	-.41
I_4^{met}	—	.16	.11	.27	.01	-.37	.05	.28		.26	-.70
I_{rest}^{met}	—	-.41	-.08	-.61	.43	.58	-.94	.16	-.31		-.02
$A(Z_1/Z_2)$	—	-.59	-.24	-.43	.18	.47	-.43	.11	-.37	.49	

In the following we treat r_{xy} and $r_{xy|z}$ as if all variables would be normally distributed, which is not true in our case, for example, not for the Z_1 values. Nevertheless we do so because the normality is not so important.[11] The meaning of the coefficients r_{xy} and $r_{xy|z}$ is then for our n value as follows: If $|r_{xy}| > 0.17$ (0.28; 0.34) or $|r_{xy|z}| > 0.17$ (0.29; 0.34), there will be a correlation between x and y with a confidence level $w > 68\%$ (90%; 95%). If $|r_{xy}| > 0.5$ or $|r_{xy|z}| > 0.5$, we shall call the correlation strong.

Table I summarizes the correlation coefficients calculated for our experiment. The values for the ionicity B are from Pauling.[12] Values with $w > 90\%$ are underlined; values for strong correlations are doubly underlined. In addition to the values of Table I we found strong partial correlations, with the R_0 dependence eliminated, between B and I_2^{met} and $I_{\mathrm{rest}}^{\mathrm{met}}$, and between Z_1 and $A(Z_1/Z_2)$, I_2^{met}, I_3^{met}, I_4^{met}, $I_{\mathrm{rest}}^{\mathrm{met}}$, and B. We also calculated all the correlations with the covalent and ionic radii, and found in the first case no significant differences from the values with the atomic radii, and in the latter case in general smaller correlations.

The strong positive correlations between Z_1 and I_2^{met} and the strong negative correlations between Z_1 and I_3^{met}, I_4^{met}, and $I_{\mathrm{rest}}^{\mathrm{met}}$ reflect quantitatively the fact that the intensity shifts to transitions between circular orbits when Z_1 increases. The partial correlations between $A(Z_1/Z_2)$ and R_0 are strong and negative, in accordance with an earlier qualitative statement concerning the metallic radii in alloys.[13] The strong positive partial correlation between $I_{\mathrm{rest}}^{\mathrm{ox}}$ and $I_{\mathrm{rest}}^{\mathrm{met}}$ points to a common reason for the enhanced (or diminished) population of low-angular-momentum states both in the oxidized element and the oxygen. This reason may be found in the shape of the slow-muon energy spectrum: A high intensity at very low energies is expected to yield an enhanced population of low-angular-momentum states, and a low intensity a diminished one.

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