Double K-shell ionization in the electron capture decay of ^{54}Mn

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The probability per K capture for double K-shell ionization in the electron capture decay of ^{54}Mn was studied by recording coincidences between Cr K x rays when the two K-shell vacancies were filled. The probability for this decay was found to be $(3.6\pm0.3)\times10^{-4}$, which is a factor of 1.5 times the theoretical prediction by Suzuki and Law, and 3.2 times that of Intemann. The energy shifts of the Cr $K\alpha^H$ and Cr $K\beta^H$ hypersatellites were found to be 254 ± 18 and 319 ± 22 eV, respectively, which are about 5% higher than the calculation of Chen, Crasemann, and Mark.

I. INTRODUCTION

In electron capture (EC) decay, double K-shell ionization occurs when one K electron is captured and the other is ejected from the atom (shakeoff, SO) or is excited to an unoccupied bound state (shakeup, SU). The combined probability for SO and SU processes for the K shell is of order 10^{-4} per K capture owing to the somewhat compensating effects of the sudden reduction in nuclear charge and the sudden disappearance of the electron shielding. The filling of the empty K shell is accompanied by the nearly simultaneous ($\sim10^{-15}$ s) emission of either two K x rays, two Auger electrons, or by one K x ray and one Auger electron. Reviews of the phenomenon have been presented by Freedman,[1] Walen and Briancon,[2] and Bambynek *et al.*[3]

Theoretical calculations for the probability per K capture for double K-shell ionization, P_{KK}, were first performed by Primakoff and Porter[4] in 1953. Additional calculations have been performed by Intemann and Pollock,[5] Intemann,[6] Stephas and Crasemann,[7] Law and Campbell,[8] and Mukoyama *et al.*[9] In the latest theory, Suzuki and Law (SL) (Ref. 10) use Dirac-Fock-Slater wave functions and self-consistent-field calculations in their description of the initial and final electron states. Based on their calculations, the probability for shakeup, P_{KK}^{SU}, is much less than the probability for shakeoff, P_{KK}^{SO}, giving $P_{KK}\cong P_{KK}^{SO}$.

Experimental values for P_{KK} are generally deduced from coincidence measurements between the K x rays emitted when the double vacancy is filled. The first x ray, a K hypersatellite (K^H; $1s^{-2}\rightarrow1s^{-1}2p^{-1}$), and the second x ray, a K satellite (K^S; $1s^{-1}2p^{-1}\rightarrow2p^{-2}$), are shifted to higher energy with respect to normal K x rays because of the additional electron vacancy. The K^H shift has been recently calculated by Chen, Crasemann, and Mark[11] and is substantially greater than the K^S shift. Resolution of the K^H x rays from the K^S and normal K x rays is possible with modern detectors and allows trustworthy values of P_{KK} to be determined for decays which may be complicated by K x rays from EC in coincidence with K x rays from internal conversion or with low energy Compton photons.

A comparison between the latest theoretical and experimental results, recently given by Schupp and Nagy,[12] showed general agreement to within a factor of 2. A plot of the ratio of experimental values to the theoretical calculations of SL, $P_{KK}(\text{expt})/P_{KK}(\text{SL})$, was given in Ref. 12 and is reproduced here in Fig. 1. When referring to the SL theory, we use their local density approximation values denoted by P(LDA) in Ref. 10. If the ^{181}W ratio at $Z=73$ is neglected, a smooth trend for the theoretical values of P_{KK} to be too small at low Z and too large at high Z is seen. Since the ^{181}W ratio may be due to its low decay energy, this investigation on ^{54}Mn was undertaken to get another experimental value at $Z=24$ to see if the smooth trend shown in Fig. 1 was really justified. (The results of this study are also shown in Fig. 1 and will be discussed later.)

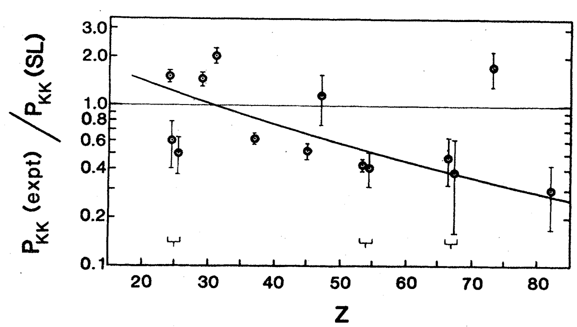

FIG. 1. Ratios of experimental values of P_{KK} to the theoretical values of SL. Isotopes for which experimental values shown are, in order of increasing Z, ^{54}Mn (this work), ^{55}Fe (Refs. 13 and 14); ^{65}Zn (Ref. 15); ^{71}Ge (Ref. 16), ^{85}Sr (Ref. 12), ^{103}Pd (Ref. 17), ^{109}Cd (Ref. 18); ^{131}Cs (Refs. 19 and 20), ^{165}Er (Refs. 21 and 19), ^{181}W (Ref. 21); and ^{207}Bi (Ref. 22). All results are plotted at the Z of the daughter atom.

The decay scheme[23] of ^{54}Mn is shown in Fig. 2 where it can be seen that the EC proceeds through the 0.835-MeV level with a half-life of 8.2 ps. This half-life is 10^3 to 10^4 times slower than electronic half-lives and thus eliminates double K-shell vacancies that could otherwise be formed by K-shell internal conversion of the 0.835-MeV transition following K capture. The probability of K^H x rays being produced by shakeoff accompanying the internal conversion of the 0.835-MeV transition is also negligible. The 312 d half-life of ^{54}Mn was convenient for the long counting times necessary in this investigation but the low energy Cr K x rays with a fluorescence yield of only 0.282 (Ref. 23) made the experiment difficult.

II. EXPERIMENTAL PROCEDURES

A. Electronic circuitry and detectors

The value for P_{KK} in this study was deduced from the coincidences recorded between the Cr K^H and K^S x rays (K^HK^S coincidences). The schematic diagram of detectors and circuits used for these measurements is shown in Fig. 3. This arrangement is very similar to that used in the ^{85}Sr investigation[12] except that the NaI(Tl) x-ray detector used in that experiment has been replaced with an ORTEC model GLP-16195/10 high purity germanium low-energy photon spectrometer, Ge(LEPS), with a 16 mm diam, 13 mm sensitive depth, 0.127 mm Be window, 5 mm window to detector distance, and peak width of 179 eV at 5.9 keV. This change was necessary to give satisfactory data for the low energy Cr K x rays. The Si(Li) x-ray detector was the same detector used in the ^{85}Sr measurements.

As can be seen in Fig. 3, two multichannel analyzers (MCA's) were used in this investigation to record pulses from the time-to-amplitude converter (TAC). A free or ungated spectrum was recorded in one MCA during the entire duration of a measurement while a second MCA simultaneously recorded a spectrum which was gated by the prompt and accidental single channel analyzers (P-SCA and A-SCA). Comparison of these spectra then allowed a determination of the TAC coincidence efficiency, E_T.

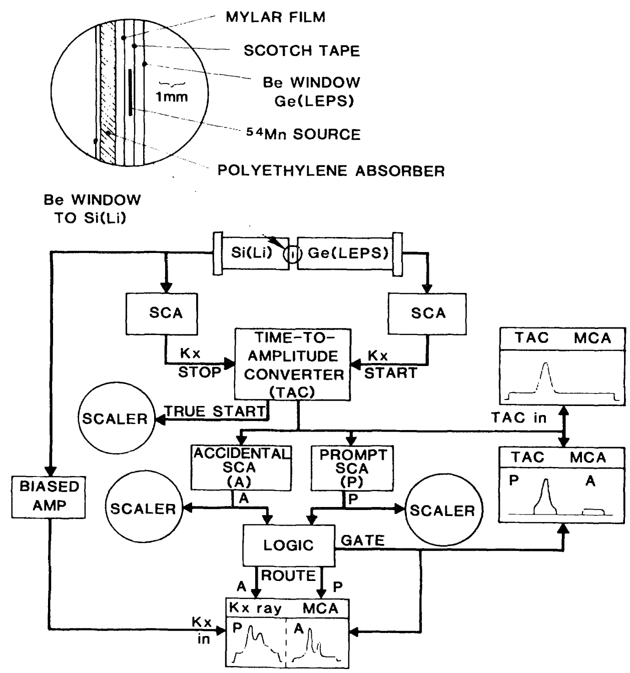

FIG. 3. Schematic diagram of detectors and circuits. Preamplifiers, amplifiers, and delay circuits have been omitted for simplicity.

B. ^{54}Mn source

The ^{54}Mn activity used in this study was purchased from the New England Nuclear Corporation and no evidence for complicating impurities was seen in a long run with a well-shielded Ge detector system at the Missouri University Research Reactor (MURR) facility. During the course of the experiment three separate ^{54}Mn sources were used. The average source strengths and duration of each run were 0.57 μCi for 335.2 h, 0.91 μCi for 319.5 h, and 1.93 μCi for 159.7 h. Each source was prepared by evaporating drops of active solution onto a 0.07 mm thick Mylar backing. After drying, the sources were covered with Scotch Brand tape and placed between the detectors in a close geometry.

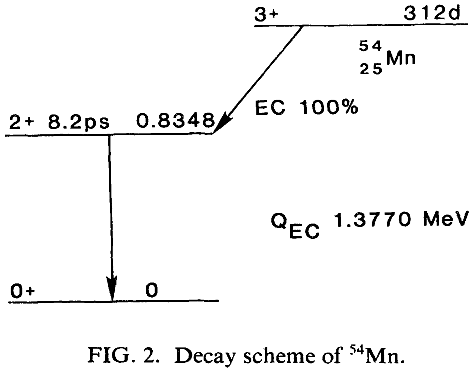

FIG. 2. Decay scheme of ^{54}Mn.

III. DATA ANALYSES

It follows from the decay scheme of Fig. 2 that the total number of K^HK^S coincidences recorded during a run, $N(K^HK^S)$, is given by

$$N(K^HK^S)=N_0(K/T)P_{KK}\omega_K^S(aE)_{\rm Ge}^S\omega_K^H(aE)_{\rm Si}^H E_T\ , \quad (1)$$

where N_0 is the total number of decays during the run, $K/T=0.911$ (Ref. 23) is the K-shell-to-total capture ratio for the EC to the 0.835-MeV level, P_{KK} is the probability for double K-shell ionization per K capture, ω_K^S and ω_K^H are the K-shell fluorescence yields for the Cr K^S and K^H x rays, $(aE)_{\rm Ge}^S$ and $(aE)_{\rm Si}^H$ are the products of absorption factors (a) and total detection efficiencies (E) for the Cr

TABLE I. Data necessary for the calculation of P_{KK} from Eq. (3).

Source (μCi)	Time (h)	$N(K^HK^S)$	$N(K)$ ($\times 10^{-6}$)	$(aE)_{\rm Si}$ ($\times 10^3$)	E_T
0.57	335.2	104.9±10.2	310.0	3.4±0.2	0.98±0.02
0.91	319.5	174.9±13.2	482.9	3.7±0.2	0.91±0.03
1.93	159.7	162.6±12.8	476.3	3.6±0.2	0.90±0.03

K^S x rays in the Ge(LEPS) and for the K^H x rays in the Si(Li) detector, and E_T is the TAC coincidence efficiency. The total number of Cr K x rays detected in the Ge(LEPS) during the same run, $N(K)$, was given by

$$N(K)=N_0(K/T)\omega_K(aE)_{\rm Ge}\,, \tag{2}$$

where ω_K is the K-shell fluorescence yield for normal Cr K x rays and $(aE)_{\rm Ge}$ is the product of the absorption and total efficiency factors for the normal K x rays in the Ge(LEPS). The very low intensity contribution to the K x rays by internal conversion as well as by the K^S and K^H x rays has been neglected in this expression. Assuming $\omega_K^S=\omega_K^H=\omega_K=0.282$ (Ref. 23) and that $(aE)_{\rm Ge}^S=(aE)_{\rm Ge}$, the ratio of Eqs. (1) and (2) gives

$$P_{KK}=\left[\frac{N(K\alpha^HK^S)}{N(K)}\right]\frac{1}{\omega_K}\frac{1}{(aE)_{\rm Si}^H}\frac{1}{E_T}\,. \tag{3}$$

The $(aE)_{\rm Si}^H$ factor in Eq. (3) was taken to be equal to $(aE)_{\rm Si}$ and was determined from both singles spectra measurements of the K x-ray count rates using the measured decay rates of the ^{54}Mn sources and from auxiliary coincidence measurements between K x rays produced in the EC decay to the 0.835-MeV excited level and the 0.835-MeV gamma rays. The values for $(aE)_{\rm Si}$ were consistent to better than 3% with respect to the average values for each run which are given in Table I.

The $N(K\alpha^HK^S)$ term of Eq. (3) was determined from least-squares computer fits made to the $K\alpha$-$K\beta^H$ region of the prompt coincidence spectra recorded with the Si(Li) detector. For a given run, the $K\alpha$-$K\beta$ region of the accidental spectrum was first fit with the sum of a modified Gaussian distribution, one representing the $K\alpha$ x rays and one representing the $K\beta$ x rays, and a linear continuum. The modified Gaussian distributions used had the form suggested by Jorch and Campbell.[24] This fitting of the accidental spectrum was done to determine those parameters which determine the shape of the $K\alpha$ and of the $K\beta$ x-ray peaks. The shape parameters for the $K\alpha$ x-ray peak determined in this manner were then used as the shape parameters for the $K\alpha$ and $K\alpha^H$ x-ray peaks, and those determined for the $K\beta$ x-ray peak were used for the $K\beta$

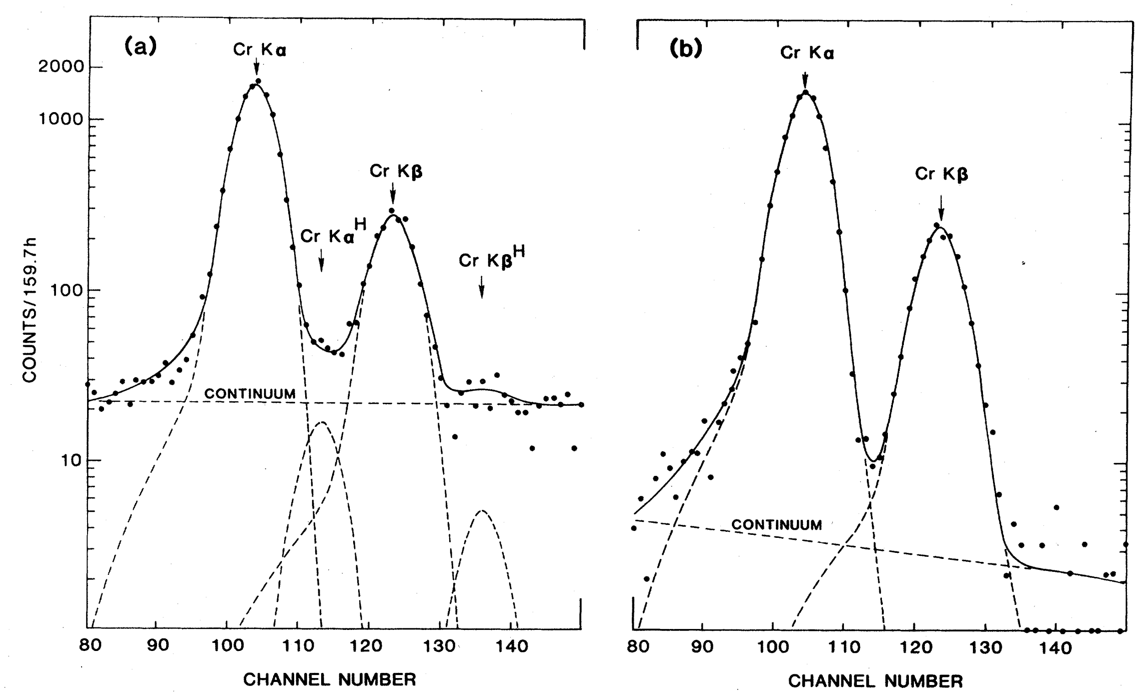

FIG. 4. (a) Prompt coincidence spectrum in the $K\alpha$-$K\beta^H$ region. The solid curve gives the overall fit to the data, whereas the dashed curves give the components of this fit, the $K\alpha$, $K\alpha^H$, $K\beta$, and $K\beta^H$ peaks, and the linear continuum. (b) Accidental coincidence spectrum in the $K\alpha$-$K\beta$ region.

TABLE II. Results of this investigation for P_{KK}, $\Delta E(K\alpha^H)$, $\Delta E(K\beta^H)$, and $K\alpha^H/K_T^H$.

Source (μCi)	P_{KK} ($\times 10^4$)	$\Delta E(K\alpha^H)$ (eV)	$\Delta E(K\beta^H)$ (eV)	$K\alpha^H/K_T^H$
0.57	3.6±0.4	259±18	279±20	0.67±0.05
0.91	3.8±0.4	239±17	341±24	0.76±0.03
1.93	3.3±0.3	263±18	338±24	0.76±0.04
Average	3.6±0.3	254±18	319±22	0.73±0.03

and $K\beta^H$ x-ray peaks in the prompt coincidence spectrum. This prompt spectrum was then fit with the sum of four modified Gaussian distributions ($K\alpha$, $K\alpha^H$, $K\beta$, $K\beta^H$) and a linear continuum. Values for $N(K^HK^S)$ were also determined from computer fits to the true coincidence spectra obtained by subtraction of the computer fitted accidental spectra from the actual prompt spectra. These values were essentially the same as from the prompt spectra and showed how insensitive the P_{KK} determination was to experimental detail not represented in Eq. (3).

IV. RESULTS

Figure 4 shows the (a) prompt and (b) accidental coincidences recorded in the Cr $K\alpha$-$K\beta^H$ region of the Si(Li) detector for the 159.7 h, 1.93 μCi run. The solid curves indicate the overall least-squares computer fits made to the region, whereas the dashed curves indicate the components of these computer fits. The large number of both $K\alpha$ x rays and $K\beta$ x rays seen in the (a) prompt spectrum are due mainly to accidental coincidences. However, several other sources of true K x rays present in these peaks are (i) K^S x rays in coincidence with K^H x rays in the Ge(LEPS); (ii) K x rays in coincidence with K x rays in the Ge(LEPS) produced by coincidences between EC K x rays and K x rays from the K internal conversion of the 0.835-MeV transition; (iii) K x rays following EC in coincidence with internal bremsstrahlung photons produced in the EC decay and detected in the Ge(LEPS); and (iv) K x rays following K EC in coincidence with the low-energy Compton distribution of the 0.835-MeV gamma rays in the Ge(LEPS).

Table I shows the data needed to calculate P_{KK} from Eq. (3) for each run. Table II shows the results of these calculations as well as an average P_{KK} value. The uncertainties on the individual and average P_{KK} values were based on a statistical contribution from the total number of K^H coincidences represented folded with independent systematic uncertainties for the $(aE)_{\text{Si}}$ factor and the E_T factor. The average P_{KK} value of $(3.6\pm0.3)\times10^{-4}$ is a factor of 1.5 times the SL theory[25] and a factor of 3.2 times the Intemann theory.[26]

Also shown in Table II are the energy difference between the $K\alpha$ and $K\alpha^H$ x rays, $\Delta E(K\alpha^H)=E(K\alpha^H)-E(K\alpha)$; the energy difference between the $K\beta$ and $K\beta^H$ x rays, $\Delta E(K\beta^H)=E(K\beta^H)-E(K\beta)$; and the $K\alpha^H$ to K_T^H ratio, $K\alpha^H/K_T^H$. The uncertainties for the individual energy differences were estimated to be about 7% from the fitting variations. This value is consistent with that obtained from deviations about the mean, but the energy resolution of the hypersatellite did not warrant a smaller uncertainty for the averaged value. The average values of both $\Delta E(K\alpha^H)$ and $\Delta E(K\beta^H)$ are about 5% larger than predicted by Chen *et al.*[11]

For normal Cr x rays, $(K\alpha/K_T)=0.88$.[23] Åberg *et al.*[27] have pointed out, however, that the ratio of $K\alpha_1^H$ to $K\alpha_2^H$ decreases considerably at low Z. Normal Cr K x rays have a value of 1.98 for this ratio, whereas Åberg *et al.* have estimated $(K\alpha_1^H/K\alpha_2^H)$ to be near zero for $Z=24$. Assuming only the $K\alpha_1^H$ transition to be suppressed yields $(K\alpha^H/K_T^H)=0.71$, a value in good agreement with the average value of (0.73 ± 0.03) shown in Table II.

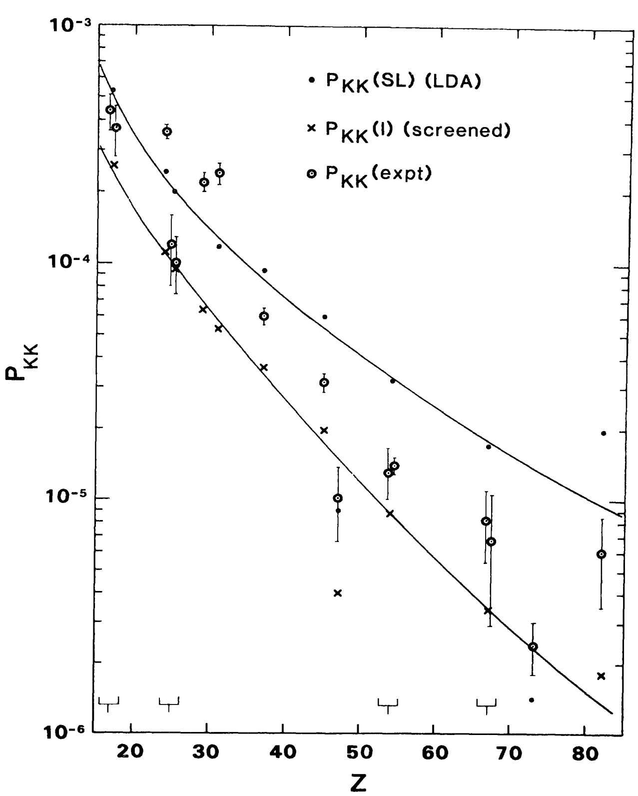

FIG. 5. Comparison of P_{KK}(expt) values with the theoretical calculations of SL and I. In addition to the recent experimental values referred to in Fig. 1, the values for ^{37}Ar (Refs. 28 and 29) are included for completeness. The smoothed curves through the theoretical values were drawn only to aid the eye.

V. DISCUSSION

As already shown in Fig. 1, the P_{KK} value determined in this investigation is consistent with the trend noted earlier for the SL theoretical values to be too small at low Z. Although the $P_{KK}(\text{expt})/P_{KK}(\text{SL})$ ratio ranges from 2.0 to 0.3, it has an unweighted average of 0.95.

Intemann (I) has recently evaluated his semirelativistic theory[6] with some minor refinements and has also investigated screening effects.[26] The results for screening with $Z_{\text{eff}}=Z-0.5$ are 22% higher than his unscreened results for ^{37}Ar and smoothly decrease to 6% higher for ^{207}Bi. When the ratio of $P_{KK}(\text{expt})$ to the screened values of Intemann,[26] $P_{KK}(\text{I})$, is plotted in the manner of Fig. 1, no significant trend with Z is seen. In this case, however, the $P_{KK}(\text{expt})/P_{KK}(\text{I})$ ratio ranges from 6.7 to 1.2 and has an unweighted average of 2.79.

Another way to present the current status between experiment and theory for SO with EC is shown in Fig. 5 where it is seen that seven of the twelve experimental values lie between the SL theory on the high side and the I theory on the low side. The smooth curves through the theoretical values have been drawn to aid the eye and help demonstrate the preceding observation. The theoretical values for ^{109}Cd and ^{181}W fall factors of 5 and 10, respectively, below the smooth curves drawn through the other values. These nuclides have significantly lower decay energy to binding energy ratios than the other nuclides and the reduced values are caused by the smaller phase space available. While the $P_{KK}(\text{expt})$ values for ^{109}Cd and ^{181}W are two of the five not bracketed by SL and I, the uncertainties permit "bracketing" within 2σ.

The three remaining $P_{KK}(\text{expt})$ values which fall above the SL theory are ^{54}Mn (this work), ^{65}Zn, and ^{71}Ge. Since we made two of these measurements, we have considered the many details of the experiments and view them completely consistent with our ^{85}Sr measurement whose $P_{KK}(\text{expt})$ value falls midway between SL and I.

In viewing the relative strengths and weaknesses of the SL and I theoretical formulations, it would seem that SL with their self-consistent-field method have treated the outer electrons very well but have not adequately accounted for the correlations between the K-shell electrons. Just the opposite statements can be made for I and his propagator approach. Perhaps some combination of these formulations would lead to a more satisfactory description of the P_{KK} process and point to further experimental investigations.

We want to thank the MURR facility and staff for preparing and calibrating the sources used, Dr. Law for supplying the $P_{KK}(\text{SL})$ value for ^{54}Mn, and Dr. Intemann for sharing with us the results of his latest calculations.

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