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CORRECTION FOR ELECTRON ESCAPE FROM SELF-SCINTILLATING CRYSTALS WITH APPLICATIONS TO THE BETA-DECAY OF K^{40}

Thesis for the Degree of M. S.
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David Goss

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ABSTRACT

CORRECTION FOR ELECTRON ESCAPE FROM SELF-SCINTILLATING CRYSTALS WITH APPLICATIONS TO THE BETA-DECAY OF K^{40}

by David Goss

The correction due to electron escape from a self-scintillating KI(Tl) 1 1/2" x 1/4" crystal was studied by the coincidence method, and compared with that calculated using the method of der Mateosian and Smith. The correction was seen to be approximately half as large as that of der Mateosian and Smith, and slightly different in form.

This correction was applied to the beta minus spectrum of K^{40}, to determine the extent of the linearity of the third order forbidden Fermi-Kurie plot. The plot thus obtained deviated from linearity at approximately 250 kev.

The empirical correction of Langer was applied to the spectrum, and the resultant F-K plot was linear to less than 100 kev.

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By

DAVID GOSS

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I also wish to thank the many people in the Department of Physics who have given advice or criticism on the various phases of the project.

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THE FERMI-KURIE PLOT

The mechanism of beta-decay is customarily explained (1) by assuming an interaction of very short range (on the order of nuclear dimensions) to exist between nucleons, electrons, and neutrinos which can cause the transformation of a neutron into a proton, or vice versa, with the simultaneous emission (or absorption) of a positive or negative electron and a neutrino. The interaction is not to depend explicitly on the linear momentum or the orbital angular momentum of the particles, but may depend on their spin states, and will depend implicitly on the angular momenta through the shape of the wave functions. An implicit particle velocity dependence may be required by the necessity of relativistic invariance. The interaction must also be completely symmetric in emission and absorption and in the sign of the charge, provided energy and total charge are conserved in each transition. Then the transition probability per unit time will be proportional to the probability of finding the particles in the region of interaction and the density of states per unit energy interval available to the emitted particle.

Consider the emission of an electron with total energy W (in units of mc^2) and momentum of $p = (W^2 - 1)^{1/2}$ accompanied by a neutrino of energy K and momentum q. The probability of the transition occurring per unit time is then

[1]* $P(W)\,dW = \text{constant} \left|M\right|^2 pWqK\,dW = \text{constant} \left|M\right|^2 pW(W_m - W)^2\,dW$

This equation assumes zero neutrino rest mass. In relativistic units $q = K = W_m - W$. W_m represents the end-point energy of the beta spectrum. $\left|M\right|^2$ is the square of the nuclear matrix element for the transition, and measures the overlap of the initial and final states of the transformed nucleon. $pW(W_m - W)^2$ is the volume of phase space into which the decay may lead. The constant term may contain the Fermi function $F(Z, W)$ which is the square of the relativistic Coulomb wave functions of the electron or positron evaluated at the nucleus, as well as a factor giving the strength of the interaction.

If the number of particles per energy interval per unit time is observed, appropriate normalization will yield $P(E)\,dE$, where E is the observed (kinetic) energy. Since $W = 1 + \frac{E}{mc^2}$, we may conclude that $P(E)\,dE = P(W)\,dW$. We may now arrange our earlier results to give

[2] $$\left[\frac{P(E)\,dE}{a_n F(Z, W)\,pW}\right]^{1/2} = \text{constant}\,(E_m - E)$$

Hence, if the left side of the above equation is plotted as a function of the beta-particle energy, a straight line should result. Such a graph is called a Fermi-Kurie plot (hereafter referred to as a F-K plot). a_n is a factor depending on the likelihood of the transition, and its usage will be described later.

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*This equation may also be written in terms of the momentum,

$P(p)\,dp = \text{constant}\,(W_m - W)^2 p^2 dp = \text{constant}\left[(p_m^2 + 1)^{1/2} - (p^2 - 1)^{1/2}\right] p^2 dp$

[Eq. 1'] in relativistic units.

However, owing to the fact that several types of interactions are possible within the framework already discussed, certain nuclei have more highly favored transitions than those of other nuclei. Selection rules for the interactions are based on an expansion of the matrix elements of the transition in terms of powers of R/λ (where R is a length the order of nuclear dimensions, λ the wavelength of the leptons). Transitions that contradict the zero-order selections rules are called forbidden transitions. The degree of forbiddenness is the same as the order of the expansion (in R/λ) necessary to describe a particular transition.

The shape of the spectra of the forbidden transitions deviate somewhat from the shape of the allowed spectra, for which the constant a_n (in Eq. 2) equals one. For the forbidden transitions, a_n is a function of the energy of the decay products, and the nuclear matrix elements, and this must be taken into account in order to obtain a linear F - K plot.

If the spin change in the beta-decay transition is a maximum for a definite order of forbiddenness, the transition will show a spectrum shape that is independent of the nuclear structure, and depends in an unambiguous fashion upon the energy (i. e. a_n depends only on the energy of the decay products). Such a spectrum is called a unique shape spectrum. For lower spin changes the spectral shape depends on various ratios of the different nuclear matrix elements. These spectra are termed mixed shape spectra.

The shape of a forbidden spectrum is more sensitive to the variation in the parameters of the theory of beta-decay than an allowed spectrum.

Furthermore, the existence of the unique shape spectra is one of the strongest legs of evidence for that class of interactions known as Gamow-Teller interactions.

The beta-decay (negatron) of K^{40} is third order forbidden, the highest order unique forbidden beta-decay yet discovered, with a spin change of four units and a change of parity (2). Because of the relatively small nuclear charge ($Z = 19$), Coulomb effects are not important, and the third order forbidden correction factor may be written as

$a_3 = p^6 + q^6 + 7p^2q^2(p^2 + q^2)$ (2).

Hence, it is anticipated that a plot of

$$\left[\frac{N(E)}{a_3 PW\, F(Z, W)}\right]^{1/2} = \left[\frac{N(E)}{f_3}\right]^{1/2}$$

as a function of the electron energy E would yield a straight line for this spectrum. N(E) is the number of transitions per unit time for a given energy and particular sample of the emitter, and is thus related to P(E), the transition probability per unit time, by a constant.

Many measurements (3, 4) of the beta spectrum of K^{40} have yielded F-K plots which deviate from linearity in the low-energy region quite significantly. This deviation has been attributed to the finite source thickness of sources used in magnetic focusing spectrometers. Since K^{40} has a half-life of approximately 1.3×10^9 years and forms only 0.119% of natural potassium (5) rather thick sources have had to be used. This difficulty may be circumvented by making the source internal to the detection system of the beta spectrometer by using a crystal containing

K^{40} in a scintillation spectrometer.

Beard and Kelly (6) showed that it was possible to linearize the K^{40} plot to approximately 150 kev by assuming a relatively large correction for electron escape from the self-scintillating crystal. It was therefore decided to determine the correction experimentally, compare with the theory, and find out whether the F - K plot was actually linear at low energies.

Langer et al. (7, 8) have found low-energy deviations in the F - K plots of several spectra. The deviations were shown to be non-instrumental in origin, and all could be eliminated by multiplying the N(E) obtained by an energy-dependent factor which was of the same form for each plot. Langer's results provided an additional point of interest to this investigation, since they brought up another factor to be considered in determining the extent of linearity of the K^{40} F - K plot.

II. THE CALCULATION OF ELECTRON ESCAPE

Studies of the beta-decay spectrum of K^{40} by scintillation spectrometer may be placed in two categories, those in which the source is external to the detecting crystal (generally assumed to have approximately 2π geometry), and those in which the source is distributed fairly uniformly within the crystal (4π geometry). The former method has the disadvantage of distorting the spectrum studied, due to the betas losing energy before scintillating by traversing a finite source thickness. A drawback to the second method is the escape of a fraction of the betas from the crystal before losing all their energy by scintillation.

The correction for electron escape from self-scintillating crystals (or organic solvents) is due to der Mateosian and Smith (9). The assumptions made in their treatment of the problem are that the electron travels in a straight line while in the crystal a distance equal to its range, until it either loses all its energy by scintillation or escapes; and that the electron loses its energy uniformly along the path. The development of the correction is as follows: If an electron of initial energy E_1 escapes the crystal before giving up all its energy, the corresponding pulse produced by the detection system will represent an energy E_2, where E_1 is greater than E_2. Let the range of electrons of energy E_1 be R_1, the range of those with energy E_2 be R_2. Further, let $N_1\,dV$ be the number of electrons of energy E_1 produced in volume

dV per unit time. If YZ (see Fig. I) represents the crystal's surface, electrons of energy E_1 coming from the volume dV at a depth x less than R_2 which escape from the surface in a ring of radius $R_2 \sin\phi$ will produce scintillations of apparent energy E_2. The solid angle (infinitesimal) presented by this ring to the element of volume dV is

$$d\Omega = \int_0^{2\pi} \frac{(R_2)^2}{(R_2)^2} \sin\phi \, d\phi \, d\theta = 2\pi \sin\phi \, d\phi:$$

then

$$\frac{N_1 dV}{4\pi} \, 2\pi \sin\phi \, d\phi = 1/2 \, N_1 \sin\phi \, d\phi \, dV$$

is the number of pulses per unit time of energy E_2 due to electrons of initial energy E_1 emerging from the volume element dV. By this token, the differential of the number of pulses of energy E_2 or less due to electrons of initial energy E_1 in volume of integration is

$$dN_2 = \int_0^{R_2} 1/2 \, N_1 \sin\phi \, d\phi \, dx.$$

Note that $x/R_2 = -\cos\phi$ and $-x/(R_2)^2 dR_2 = \sin\phi \, d\phi$. Then,

$$dN_2 = \int_0^{R_2} 1/2 \, N_1 \, x/(R_2)^2 \, dR_2 \, dx = 1/4 \, N_1 \, dR_2.$$

To find the total number of electrons recorded as having energy E_2 per surface area of crystal integrate the above expression over R_2 from 0 to R_1 and obtain

$$N_2(E_2) = \int_0^{R_1} 1/4 \, N_1 \, dR_2 = 1/4 \, N_1 R_1.$$

Hence, the electrons of energy E_1 are degraded equally into all energies below E_1, since the above results are explicitly independent of E_1 and E_2; and the fraction of electrons of energy E_1 which escape from the crystal equals one-fourth the fraction of the volume of the crystal which lies in a surface layer of depth R_1.

The correction might be applied to a given spectrum by subtracting the number of counts corresponding to degraded pulses for each energy interval from the total number of counts in that interval (provided one is not interested in determining the "true" sample specific activity per given energy range, but only the spectral shape, as is the case here).

Since the range is a known function of the energy (10), the fraction of electrons degraded <u>from</u> any particular energy interval may be easily calculated and multiplied times the activity for that interval to find the number of electrons that are degraded from a particular interval. The resultant number of counts (pulses) may then be distributed equally into each lower energy interval, and the results subtracted from the original spectrum. This would then leave just the corrected spectrum.

III. THE COINCIDENCE CORRECTION

In principle, the number of pulses degraded by electron escape can be determined experimentally. This can be done by placing a NaI (Tl) crystal in contact with the KI(Tl) source crystal so that those betas escaping the KI(Tl) crystal would be detected. An experiment of this nature was performed. A thin, light-tight, aluminized mylar film was placed between the opposing faces of the crystals, to separate the light pulses and transmit most of the betas which would normally escape from that surface of the source crystal. The remaining face of each crystal was optically coupled with a high-viscosity silicon fluid to a 6342 photomultiplier tube, forming an end-to-end arrangement (Fig. II). Aluminum foil was the light reflector used on the sides of the crystals. Both crystals were of the same size and shape (cylinders, 1 1/2 inches diameter by 1/4 inch high).

Each photomultiplier tube was mounted on a special low-noise pre-amplifier with an ungrounded filament supply, with the output going to a fast, linear, pulse amplifier with excellent non-overload properties. The amplified output of the KI(Tl) detector (detector 2) led directly to a 256 channel pulse height analyzer. The discriminator output of the amplifier for the detector recording the escape electrons (detector 1), and the output of one from detector 2 were taken to a coincidence circuit in such a manner that the analyzer would store a pulse whenever the two outputs were in coincidence. This coincidence would thus be achieved

whenever an electron escaped from the source crystal and entered the other crystal, provided it produced scintillations in both crystals (Fig. III).

The basic idea underlying the use of the coincidence spectrum is as follows: if the count rate of pulses which are degraded from higher energies is known for each energy interval (channel), this rate subtracted from the measured rate for each energy interval should yield the spectrum corrected for electron escape. The coincidence apparatus essentially measures the correction for escape from one surface of the crystal, so that the total correction is equal to the measured coincidence rate for each channel times the ratio of the total surface area to the area of the side used.

The coincidence circuit used to unblank the 256 channel analyzer had a resolving time of approximately 7.8 microseconds, determined by using the method of independent sources. Because of this relatively large resolving time, the difference in the rise times characteristic of the NaI(Tl) (approximately 0.25 microsecond) and KI(Tl) (approximately 1.0 microsecond) crystals had little influence on the triggering of the coincidence circuit. The accidental rate was very small (less than 0.1% of the coincidence correction in each channel) due to the low count rate in each detector. The final results were corrected by an almost negligible amount to account for the accidental counts.

The experimental correction for electron escape to be applied to the K^{40} beta spectrum is shown in Fig. IV, along with the theoretical

correction predicted by der Mateosian and Smith. The theoretical curve was computed from the spectrum of the 1 1/2" X 1/4" KI(Tl) crystal used in the experiment. A suggestion due to G. B. Beard (11) states that since only half of the electrons survive the distance commonly denoted by the range (i.e., the mean range; this is the range referred to as a known function of the energy), the above theoretical correction should be multiplied by a factor of 1/2. This type of correction is also shown in Fig. IV. The experimental correction is of approximately the same shape as the der Mateosian-Smith correction, and corresponds roughly to the results predicted by Beard.

The actual coincidence runs all showed a sharp cut-off at the low-energy end of the spectrum. This is attributed to the stopping and back-scattering of low energy betas by the reflected film. This film was doubly aluminum-coated mylar,[1] of measured thickness 1.1 $\pm$ 0.3 milligrams/cm^2. Calculations based on the known range of electrons in matter (see above) (10) and an estimate of effective film thickness as a function of the angle of incidence (assuming the betas travel in straight lines through the film) showed that the stopping energy was less than 25 kev for betas striking the film at an angle to the normal of less than 60^o.

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[1]Several zapon and collodian films coated with aluminum by vacuum deposition (thickness 0.18 - 0.06 mgm/cm^2) were tried in this capacity, but such films seldom had the tensile strength to withstand the strain of mounting. Once mounted, these films were found to give light leaks between the two detectors, and so were abandoned. The light leaks were detected by noting pulses of two different rise times at the output of the preamplifier, corresponding to the different rise times of the NaI(Tl) and KI(Tl) crystals. The corresponding output using the mylar film did not show this discrepancy.

At angles less than 87^{o}, the stopping energy was still less than 100 kev. Considering the small percentage of electrons escaping into the escape detector with an incidence angle greater than 87^{o}, the effect of film thickness should not noticeably affect the escape correction shape for energies greater than 100 - 120 kev.

To account for electrons which escaped through the common surface between detectors, but for some reason did not trigger the escape detector, we shall assume an error in the coincidence spectrum proportional to the number of counts per given energy range. This amounts to assuming that a constant fraction of the electrons for each energy interval which escape bearing an undetermined energy are stopped in the film.

The above assumption indicates that the escape spectrum should be multiplied by a factor of (1 + f) in order to account for the effect of finite film thickness, where f is the fraction of electrons escaping in the crystal at the film interface which do not trigger the escape detector. f may be roughly estimated by taking the ratio of the number of electrons with energy less than an energy E_1 which occur on the escape detector's integral spectrum, to the number of the total number of escape electrons actually detected. E_1 is the threshold energy of the escape detector. In view of the results of the stopping energy as a function of angle for the mylar film, this energy was taken to be approximately 60 kev. The integral spectrum measured by the escape detector was extrapolated from this energy to zero energy. From these results it was found that an error of 2.8% is made in the escape spectrum by neglecting f. This becomes negligible when applied to the beta spectrum as a whole.

As a check on these measurements, the measured coincidence spectrum was extrapolated to zero thickness of the partitioning film by taking the coincidence measurements with various thicknesses (i.e., separate layers) of the reflecting film. All these measurements agreed within experimental error (approximately 3% over most of the energy range) above 120 kev. The extrapolation is only slightly greater than the one-layer results at the low energies above cut-off. In conclusion, it seems that the distributed error due to finite film thickness is best corrected at low energies by extrapolation of the experimental curve to zero energy, and may safely be neglected over most of the rest of the spectrum.

Since the edges of the escape detection crystal do not subtend a 2π solid angle to some nuclei in the source crystal, the effect of electron escape without detection from the edge of the source crystal was also taken into account. An evaluation of the crystal efficiency leads to an upper limit of about 3% for this effect; however, the results of the coincidence runs with varying film thickness indicate that this effect, too, is negligible.

The coincidence spectrum results were checked by comparison with the difference of a beta spectrum and an anti-coincidence spectrum. The anti-coincidence circuit was adjusted in such a way that it effectively "blanked" the 256-channel analyzer when the pulses from detectors 1 and 2 were in coincidence. The results obtained by the difference method are the same as the regular coincidence results, although not as reliable

statistically, since they constitute a small difference between two large numbers, and are comparable in magnitude to the uncertainty in either. For this reason the difference method was not exploited fully to correct the beta spectrum.

The error in the various points in the coincidence spectrum is denoted by the regularly spaced flags shown in Fig. IV. The large error at the very low energy end of the spectrum is due mostly to the uncertain operation of the 256-channel analyzer in this region.

In conclusion, it seems that the experimental correction differs significantly from the theory at high energies and is overall a smaller correction than that predicted by der Mateosian and Smith, yet larger than that predicted by Beard.

IV. EXPERIMENTAL RESULTS

The beta spectrum of K^{40} was studied by observing the natural activity of a one inch diameter by one inch high cylindrical KI(Tl) crystal. Observations were also made with a one and one-half inch diameter by one-fourth inch high crystal, but because of the poor resolution and large electron escape associated with the crystal, the error was quite high, and the results, while they agree within the bounds of experimental error with those obtained with the larger crystal, are thought not to be as significant. The crystal used was mounted on a 6342 photomultiplier tube for use as part of a scintillation spectrometer in conjunction with the 256-channel pulse height analyzer.

The beta spectrum thus obtained is shown in Fig. V. Corrections were made to the spectrum to account for the background radiation, finite spectrometer resolution (12), and electron escape (experimental correction). The estimated errors are shown by the error flags. For points without flags, the error is smaller than the radius of the small circles circumscribing the points. The statistical error was less than 1% at any point; the large error at extreme energy ranges is due mostly to uncertainties in the background and resolution corrections. The error in the resolution correction is less than 3% over most of the spectrum, and because of the small magnitude of the correction, is nearly negligible. This correction is appreciable only at the extreme high energy end. The beta spectrum is similar in shape to that obtained by Kelly and Beard (6).

The background count was reduced to an almost negligible value over most of the energy range by shielding the detector with copper, steel, and lead. The weak (about 12% of transitions) 1.46 mev gamma ray of K^{40} was accounted for by fitting a gamma spectrum of roughly that energy (1.28 mev) obtained with an external source to the approximate location on the spectrum occupied by the 1.46 mev gamma. Because of the small crystal efficiency corresponding to such a high energy, the total activity detected is estimated at less than 2% of total beta transitions. Earlier background measurements taken with an external KI(Tl) crystal as a source (shielded from the background detector in such a manner as to eliminate most of the betas) failed to show the location of the gamma peak or the extent of its influence on the count rate of the high energy end of the beta spectrum. The background was measured in each case with a NaI(Tl) crystal of the same dimensions as the KI(Tl) crystal, and the difference in scintillation efficiencies of the two materials accounted for.

The experimental coincidence correction described earlier was also applied to the spectrum, and had the effect of shifting the beta peak slightly toward the higher energy end of the spectrum.

The F-K plot for this spectrum is shown in Fig. VI. It deviates from a straight line at approximately 250 kev. This plot is of the same form as a plot made by Beard and Kelly (13) utilizing one-half the magnitude of the der Mateosian-Smith correction. The deviation at low energies appears to be outside the range of error, as denoted by the error flags in Fig. VI.

V. CONCLUSIONS

It now appears probable that the F-K plot of K^{40} deviates from linearity at very low energies; that is, the third order forbidden shape factor is slightly smaller than it should be, or other corrections should be made that were not taken into account by this investigation. For example, it is known that KI(Tl) crystals exhibit a known delayed phosphorescence which might produce spurious low energy pulses (14); however, this effect is thought to be too small to affect the experiment (15).

Langer et al. (7, 8) have noticed some low-energy deviations from linearity of the F-K plots for several beta spectra. These deviations are thought to be non-instrumental in origin, and could be corrected by multiplying the N(E) found by $\frac{1}{1+b/W}$, where W is the total relativistic energy in units of mc^2 and b is a small constant, between 0.2 and 0.4 (mc^2) in value.

Using a value of b = 0.3, a plot of this type was constructed using the K^{40} results. This plot is nearly linear in the low-energy region to about 100 kev (Fig. VII), and all except two of the points are within the range of error of the line.

This agreement with Langer's results must be regarded merely as fortuitous in the present instance, since there are probably other factors with which one might produce a linear F-K plot, but the results are certainly interesting.[1]

[1] See also Bulletin of American Physical Society, Series II, Vol. 6 (1961), Abstracts QA6, KA6, for further discussion of Langer's correction.

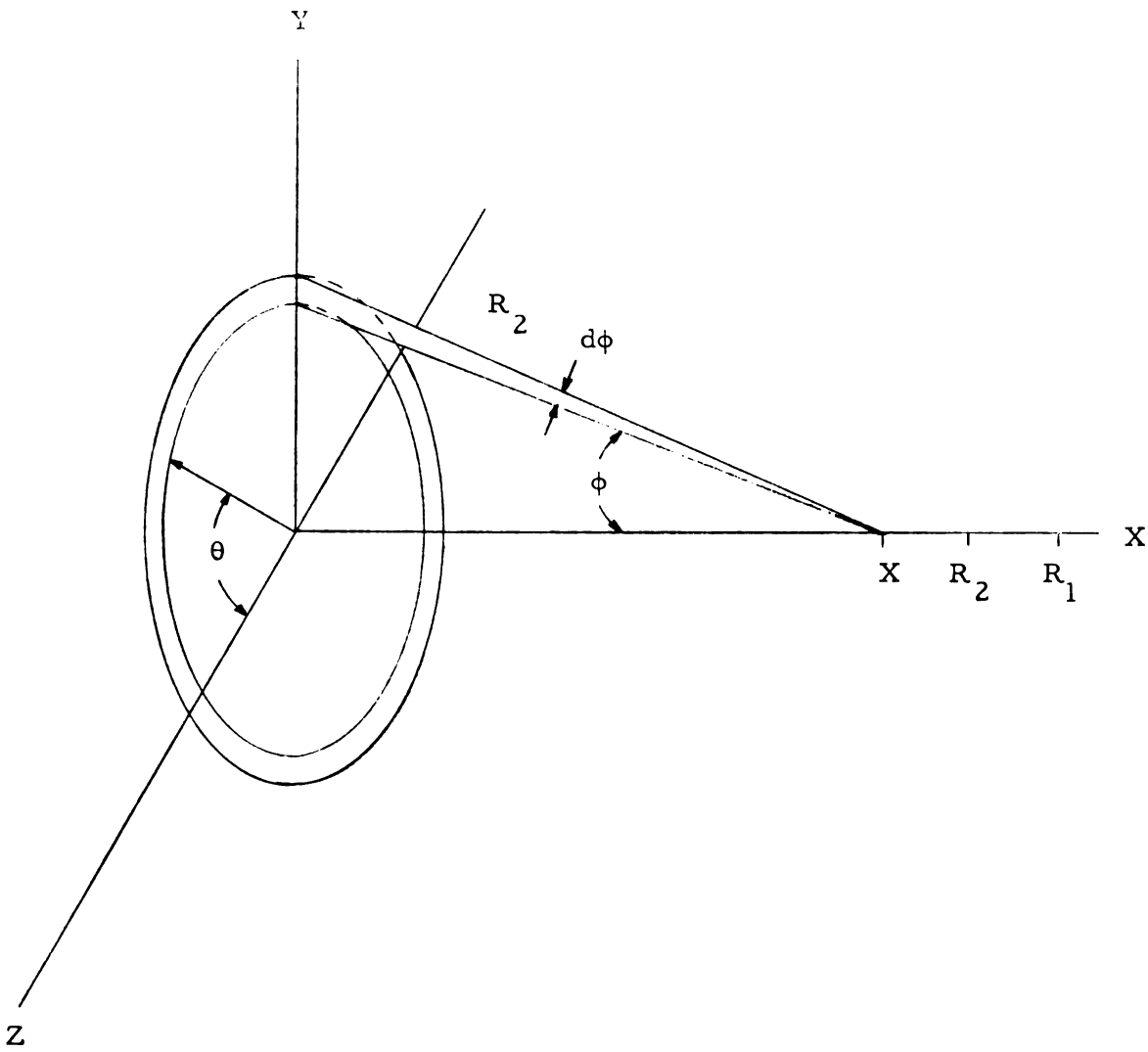

Figure I. Diagram showing the location of the parameters used in the escape correction calculations.

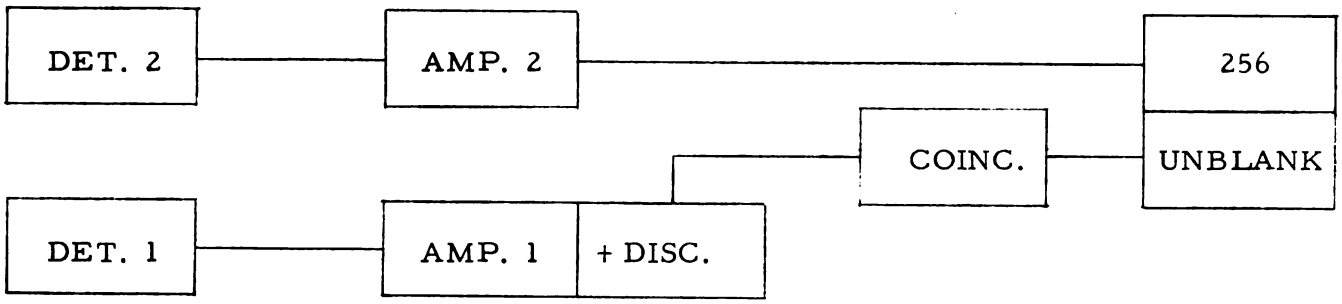

Figure III. Block diagram of circuit used to obtain coincidence correction. "Det." signifies detector, "Amp." amplifier, "+Disc." the positive discriminator output of the amplifier, "Coinc." the coincidence circuit, "256" the 256-channel pulse height analyzer.

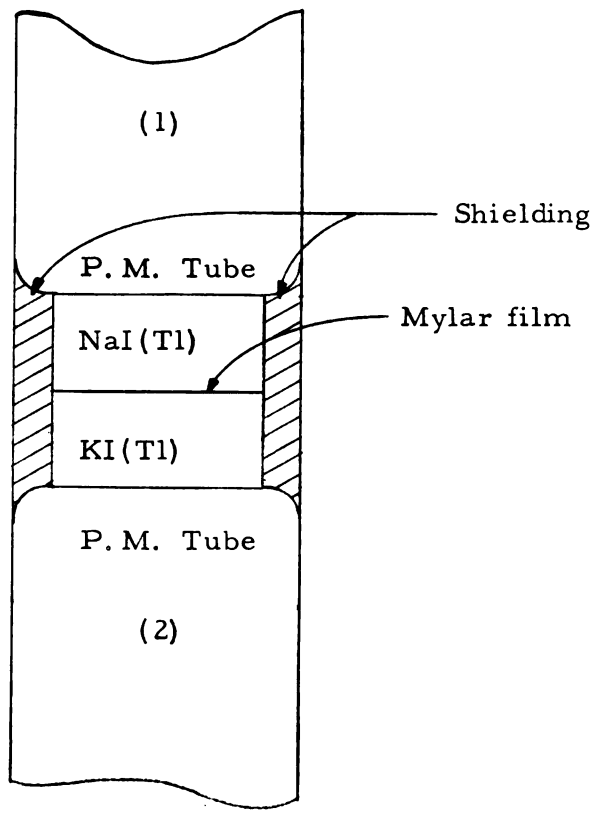

Figure II. Cross-section of the source and light-collection portion of detectors 1 and 2.

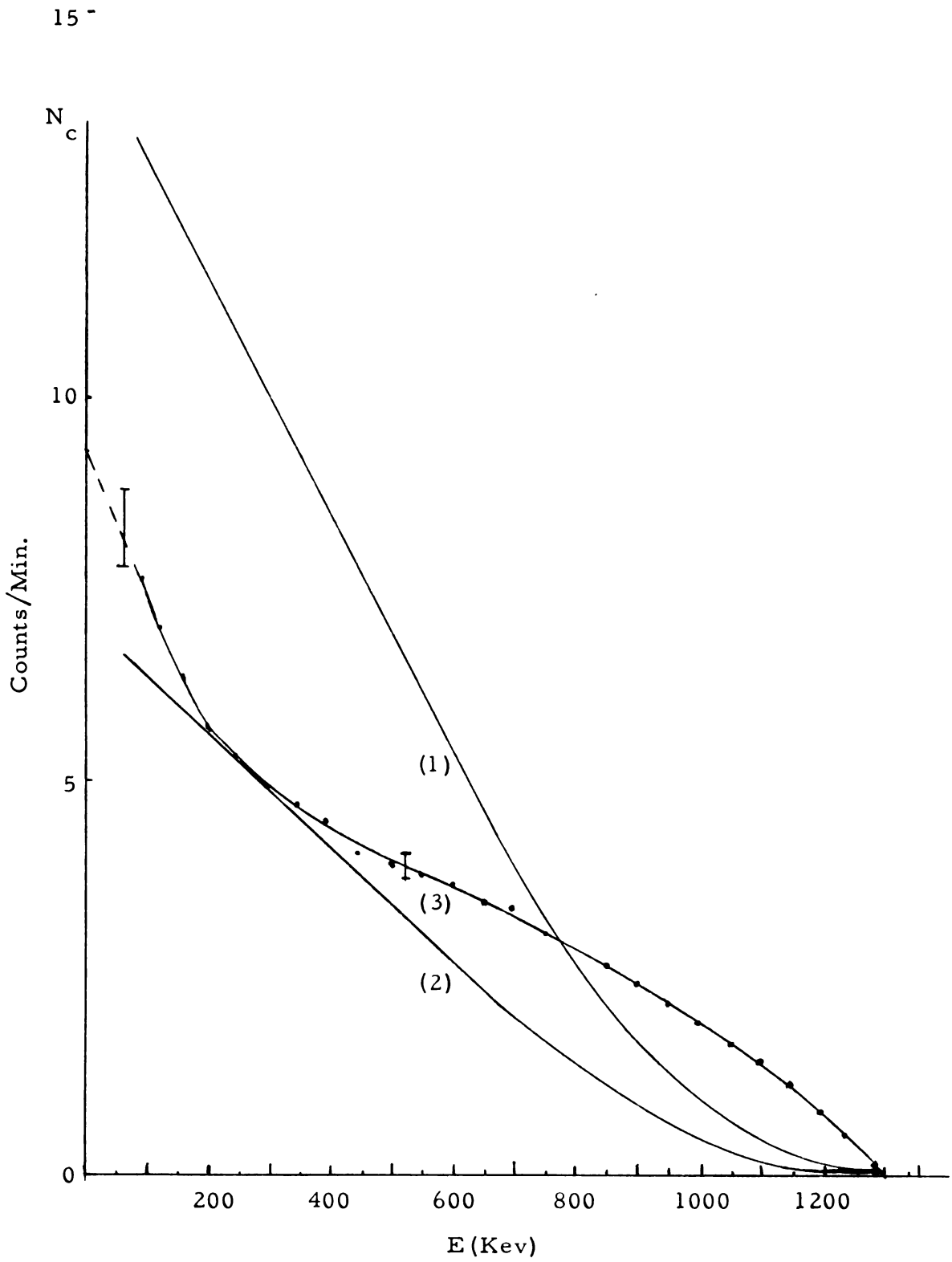

Fig. IV. (1) der M-S correction; (2) der M-S correction X 1/2;
(3) experimental correction.

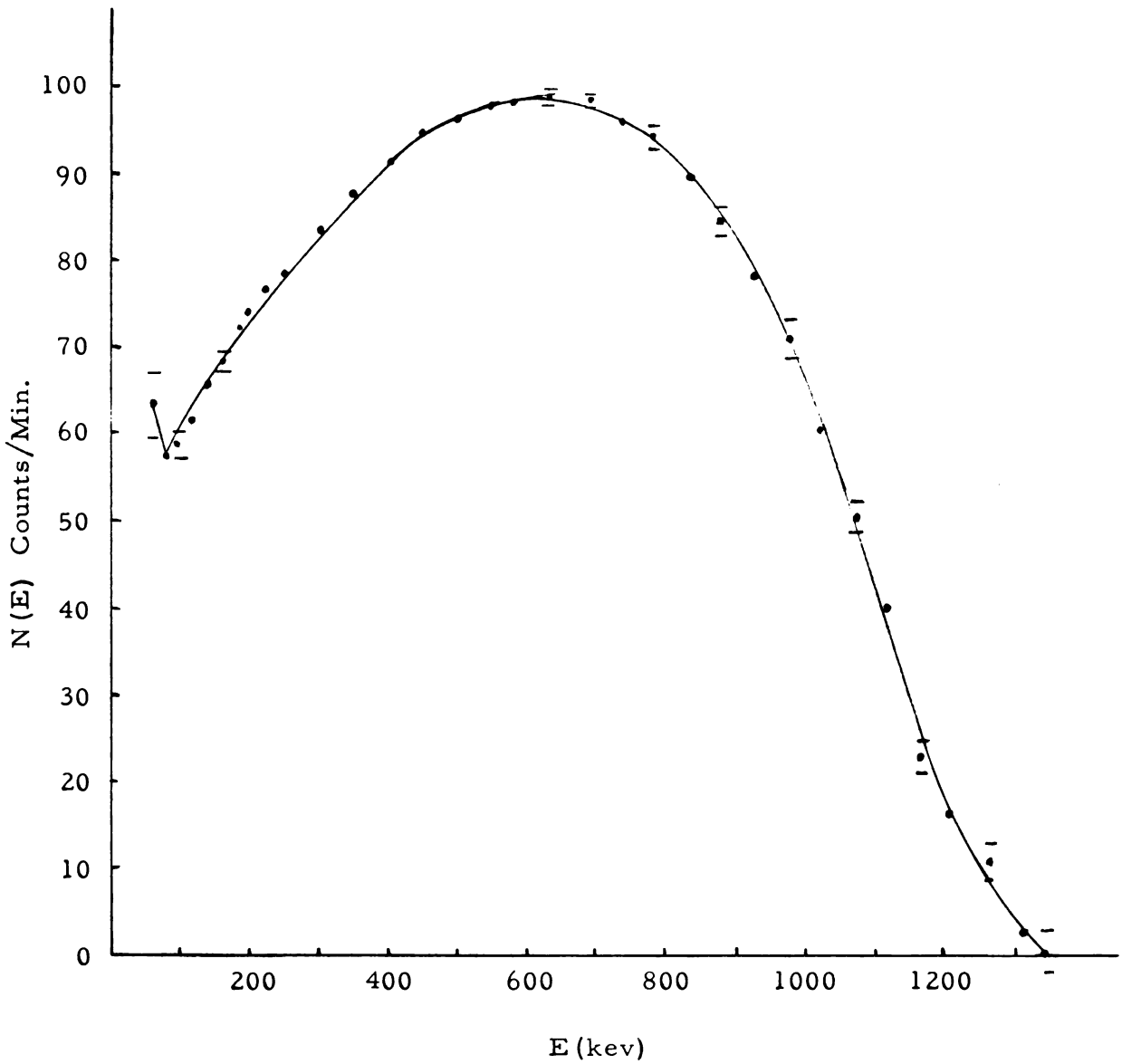

Fig. V. K^{40} B^{-} Spectrum

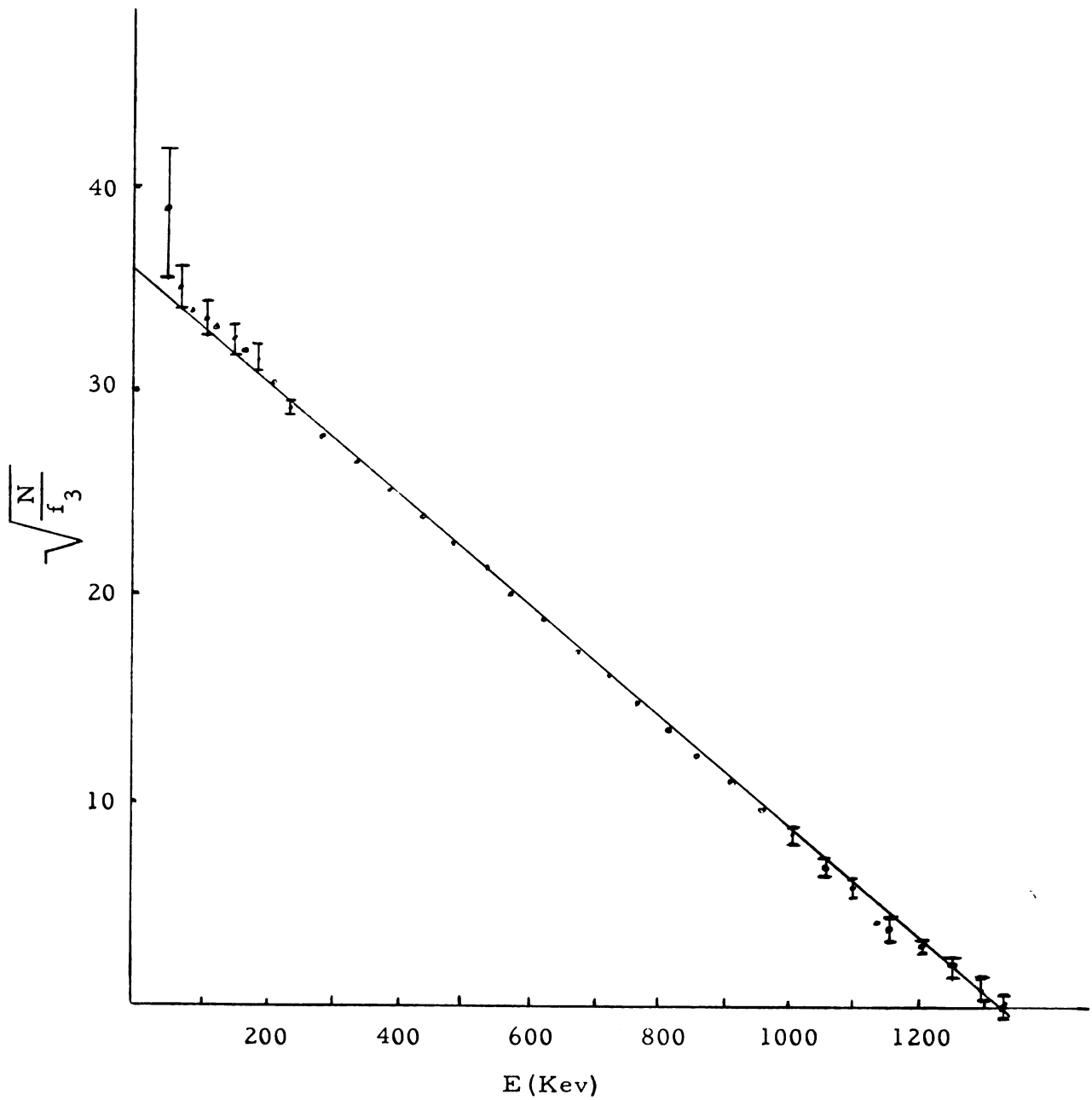

Fig. VI. F-K plot--no correction

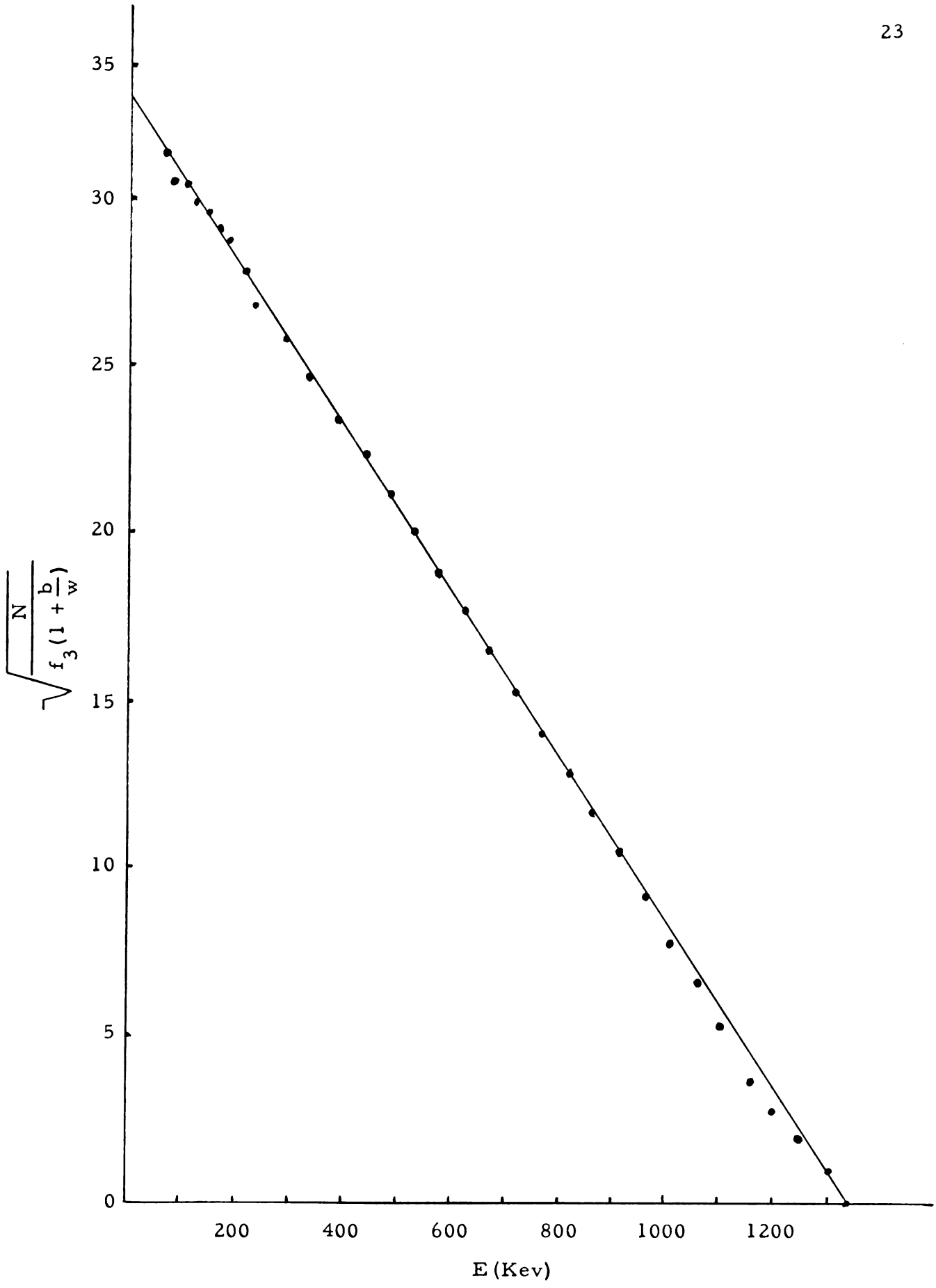

Fig. VII. Kurie plot--Langer correction

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