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1. Abstract

This paper describes the use of a high resolution beta-ray spectrometer ($\Delta p/p \sim 0.1\%$) to measure short lifetimes in nuclei recoiling from nuclear reactions or alpha decay. The method takes advantage of the Doppler shift in the momentum given to electrons emerging from recoiling atoms which is $\geq 0.1\%$ following alpha decay and $\geq 1\%$ for typical heavy ion reactions.

The 42.8 keV E2 transition in Pu^{240} following alpha decay of Cm^{244} was studied in detail as an illustrative example; the L_{II} and L_{III} conversion lines from an uncovered Cm^{244} source are shown to have the expected Doppler structure in the form of high momentum shoulders extending ~0.3% above the unshifted momentum p_o of electrons emerging from atoms at rest in the source backing. With use of an electrode system of two grids placed close to the source so that the electrons are first decelerated and then reaccelerated it is shown that the electrons emitted from recoiling atoms can be given a net acceleration which is proportional to the distance travelled by the atom from the source surface before electron emission takes place. The L_{III} 42.8 line has been studied directly and also in coincidence with alpha particles detected behind the source, with applied electric fields of up to 40,000 volts/cm. Analysis of the resulting line shapes yields a half-life of $(1.6 \pm 0.2) \times 10^{-10}$ sec, without correction for absorption in the source (which would tend to raise the value). A value of $(1.72 \pm 0.15) \times 10^{-10}$ sec has been measured electronically by Bell et al. A discussion of the potentialities of the method shows that it should be possible to measure lifetimes as short as $\sim 3 \times 10^{-12}$ sec following alpha decay and as short as 3×10^{-13} sec following reactions involving heavy ions.

AN ELECTROSTATIC METHOD FOR THE RECOIL MEASUREMENT OF SUB-NANOSECOND NUCLEAR LIFETIMES*

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2. Introduction

The determination of absolute transition probabilities of nuclear radiations is a central problem in nuclear spectroscopy, and a number of methods have been developed that are useful in different experimental situations. Recently, extensive reviews of these methods have been given by Devons[1]) and by Bashandy[2]), and, as is customary, distinction is made in these reviews between the electronic methods, applicable in general for lifetimes $\geqslant 10^{-10}$ seconds, and the various techniques suitable for shorter lifetimes. Of the latter methods, those based on Doppler or recoil effects have been perhaps the most widely used.

In essence, the recoil method seeks to determine the distance that is traversed by an atom, set in motion by recoil from a nuclear reaction or particle emission, during the lifetime of the excited nuclear state. If the recoil velocity is known, then the lifetime of the state can be calculated. The distances travelled by the recoil atoms can be measured directly by the imposition of a movable stopping foil in vacuum, or it can be inferred from the energy shift caused by passing the recoils through absorbers of varying thickness or density. As pointed out by Devons[1]), two factors severely limit the precision of these Doppler shift methods; in the first method, the necessity to perform accurate measurements of extremely small distances (10^{-3}-10^{-5} cm),

and in the second, the interpretation of results in terms of the slowing down and scattering of the recoil atoms, processes that are not adequately understood.

In this paper we propose an alternative method for measuring the distances travelled by the recoil atoms following alpha decay or nuclear reactions. Rather than perform measurements on the recoils directly, we utilize an electrostatic device that gives information about the position where a conversion electron is emitted from the moving recoil atom. In the examples to be described the Doppler effect on the conversion electron momentum is small ($\leqslant 0.3\%$), and we utilize the high resolution of an iron-free $\pi\sqrt{2}$ beta spectrometer to distinguish between the electrons emitted from the moving recoils and those emitted from atoms at rest in the source foil. The electric field is arranged so as to be effective only on the first component. From a knowledge of the atom recoil velocity and electric field strength one can interpret the change in conversion line shape to deduce the recoil distance before electron emission and hence the lifetime of the nuclear state involved.

The method is shown to be quantitative and readily applicable to lifetimes $< 10^{-9}$ seconds, but in common with all conversion electron and recoil studies it is sensitive to the thickness of the source deposit.

3. Principle of the Method

It is well established that the recoil of nuclei after heavy particle emission produces measurable changes in the energies of gamma rays or electrons emitted from the moving atoms. In the case of an internal conversion electron emitted from the recoiling atom with momentum p_o (energy E_o) in the atom frame

of reference, the momentum observed in the laboratory frame of reference is given by:

$$p = p_o + \Delta p = p_o \left[1 + v_r/v_e \cos \theta \right] \qquad (1)$$

where v_r and v_e are the respective velocities of the recoiling atom and the conversion electron, and θ is the angle of emission of the electron with respect to the recoil direction.

Consider as an example the alpha decay of the heavy nucleus $_{96}Cm^{244}$. In this case an intense alpha group, of energy 5.76 MeV, populates the first excited state of $_{94}Pu^{240}$ at 42.8 keV. This state decays by an E2 transition to the ground state, with prominent L_{II} and L_{III} conversion electrons of energies 20.6- and 24.8- keV respectively[3]. The maximum Doppler shift of the L_{III} line (at $\theta = 0^o$) is calculated from the electron velocity ($v_e = 0.30$ c) and the Pu^{240} recoil velocity ($v_r = 9.3 \times 10^{-4}$ c) to be 0.31% in momentum. Figure 1 shows this conversion line as recorded in a 50 cm $\pi\sqrt{2}$ iron free spectrometer[4,5] with an uncovered Cm^{244} source; the Doppler "shoulder" on the high energy side of the line is evident. Figure 2 shows another example of a Doppler shoulder, this one observed on the L_I line of the 41.8-keV transition following alpha decay of $_{99}Es^{253}$ [6]. In each figure the calculated maximum energy shift is indicated.

In the idealized situation of an infinitely thin source and much higher instrumental resolution one would expect half the electron intensity to be due to atoms recoiling forward from the source backing. The appropriate integration of eq. (1) would then predict a rectangular shoulder distribution in momentum extending from p_o to $p_o + \Delta p$. In practice, however, the "ideal" distribution

of a sharp line at p_o plus a rectangular high momentum shoulder is smeared by the finite instrumental resolution and source thickness effects, as seen in figs. 1 and 2. It should be noted that in the event of a nuclear lifetime short compared with the slowing-down time of the recoil atom in the material of the source backing (typically $\sim 10^{-13}$ sec.) a corresponding shoulder will appear on the low momentum side of p_o. In the cases of interest here, $\tau \gg 10^{-13}$ sec., and therefore no low momentum shoulder is distinguishable.

As mentioned above, it is possible in principle to make direct measurements of the distances travelled by recoil atoms before decay, and in fact, Burde and Cohen[7]) and Siekman and deWaard[8]) have performed ingenious determinations of this type of the half life of the 40 keV excited state in Tl^{208} following alpha decay of Bi^{212}. These measurements were made by placing a movable absorber in front of the source in a beta spectrometer and noting the energy shift of the L_I conversion line (ThB "A" line) as a function of the distance between source and absorber. Because of the low recoil velocity associated with alpha decay ($v_r \sim 10^{-3}$ c) the recoil distances in this type of measurement are around 10^{-3} - 10^{-5} cm., and quite involved physical methods must be applied if large errors are to be avoided. Also, the energy losses of the electrons in passing through the recoil-stopping absorbers add complication to the analysis of the results. It would be desirable to have an alternative means of measuring these recoil distances which is free from these difficulties and which might as a consequence have wider applicability.

We propose here an indirect but simple electrostatic method for performing the recoil-distance determination. Its essential feature is the use of an electrostatic grid system that causes a net acceleration of the conversion electrons which is proportional to the perpendicular distance

travelled by the recoil atoms from the source surface before electron emission, and thus provides a measure of that distance and therefore of the elapsed time between formation and decay of the excited state.

A diagrammatic view of the pre-accelerator system (so called because the acceleration takes place before momentum analysis of the electrons in the spectrometer) is given in fig. 3. It consists of a thin source backing on which the active material is placed, and of two metallic grids placed parallel to the source foil. The source foil and the second grid are kept at ground potential while the first grid is placed at a high negative potential; therefore the electric field in the first half of the system is opposite to that in the second half. In this arrangement an electron leaving the source foil in the direction of the spectrometer entrance baffle will be decelerated in the first half and accelerated in the second half of the device, but because the total potential drop along the electron path is zero there will be no net change in electron energy. However, if the electron emission occurs somewhere between the source and the first grid, then the energy loss due to the deceleration will be smaller than the energy gain due to the acceleration, and the electron will leave the second grid with an energy increment. This energy increment will be the greater, the farther from the source electron emission takes place, and it is proportional to that distance.

Consider the simplest case of a recoiling nucleus, left in an excited state after alpha decay, which leaves the source and travels in the direction of the spectrometer baffle ($\theta = 0^{\circ}$) with velocity v_r. The distance which the recoiling atom travels is $\ell = v_r \cdot t$, where t is the elapsed time between alpha particle emission and conversion electron emission. Obviously, from a measurement of ℓ, t can be found if v_r is known.

As the electron reaches the first grid it has energy

$$E_i - \left(1 - \frac{\ell}{L}\right) V_o \cdot e,$$

where E_i is the initial electron energy, e is the electronic charge, V_o is the applied voltage, and L is the distance from source to first grid. The electron then enters the accelerating field where it receives the energy increment $V_o \cdot e$. Therefore the total energy after leaving the second grid will be

$$E_f = E_i - \left(1 - \frac{\ell}{L}\right) V_o \cdot e + V_o \cdot e = E_i + \frac{\ell}{L} V_o \cdot e$$

or numerically equal to

$$E_f = E_i + \frac{\ell}{L} V_o \text{ (electron volts)}$$

If we denote $E_f - E_i = \epsilon$, then

$$\ell = \frac{\epsilon}{(V_o/L)} \tag{2}$$

The time t is therefore

$$t = \frac{\epsilon}{(V_o/L) \cdot V_r} \tag{3}$$

The number of electrons that receive an energy increment is proportional to the number of recoiling nuclei decaying (in a plane parallel to the source plane) at a distance ℓ. Since the time distribution of recoil atom decays is of course exponential, so also will be the energy (or momentum) distribution of the electrons emitted from in front of the source ($\ell > 0$). The expected effect of applying the voltage to the preaccelerator system is therefore to change the steep high-energy side of the conversion line into an exponential high-energy tail. The slope of this exponential tail should be proportional to the half life of the excited nuclear level $T_{1/2}$.

$$\epsilon_{1/2} \propto T_{1/2}$$

For electrons emitted from atoms recoiling forward ($\cos\theta \approx 1$) $\epsilon_{1/2}$ can be related quantitatively to the half life by eq. (3). Thus,

$$T_{1/2} = \frac{\epsilon_{1/2}}{(V_o/L)\cdot v_r} \qquad (4)$$

It should also be mentioned that the same electric field that causes deceleration of the electrons will cause acceleration of the positively charged recoil atoms. However, because of the large mass difference between the heavy recoil atom and the electron, the effect due to ion acceleration is small and can be neglected for the present purpose. For example, a heavy nucleus (A ~240) recoiling from the emission of a 6 MeV alpha particle has a kinetic energy of about 100 keV and a velocity v_r about 10^{-3} c. The overall acceleration before electron emission will be $n\cdot\epsilon$, where n is the ion charge and, since typically $\epsilon \ll 1$ keV, we see that the ion velocity will not be altered by more than ~1% before electron emission.

4. Sources and Apparatus

Sources of Cm^{244} and Es^{253} were used in this study. The Cm^{244} source was prepared by vacuum sublimation of dried chloride solution from a shaped tungsten boat at $\geqslant 2000^{\circ}$ C through a collimator 1 mm × 10 mm onto an aluminum foil backing of surface density ~3.5 mg/cm^2. The source contained about 1 μgm Cm^{244} and had a strength of 2×10^8/min. The Es^{253} source used for the data in fig. 2 was prepared in a similar fashion but without the use of a collimating slit. The active deposit had an area of about 2 mm × 10 mm, and an initial disintegration rate of $\sim 2 \times 10^9$/min i. e., contained 34×10^{-9} grams of Es^{253}.

The conversion electrons were magnetically analyzed with the Berkeley 50 cm. radius $\pi\sqrt{2}$ iron free spectrometer[4,5]). For the initial work the detector was a side window Geiger counter having a dead time of ~50 μsec. The counter window aperture, 1 mm × 20 mm, was covered with a multilayer formvar film of suface density $\leqslant$ 100 μgm/cm^2 which was > 90% transparent to the electron energies of interest. For the data shown in fig. 1 the electron baffle had an aperture solid angle of $\Omega/4\pi$ = 0.08% whereas for that in fig. 2 a larger aperture, $\Omega/4\pi$ = 0.16% was used. The focusing aberrations for these two apertures contribute widths of 0.05% and 0.1% respectively in momentum to the conversion line profile. In both cases the geometrical and electron optical factors alone would result in an overall resolution of ~0.1% in momentum. The much greater widths observed are due to the combination of source thickness and Doppler effects. The spectrometer current was programmed automatically and the output data recorded by an electric typewriter.

An exploded view of the elements of the pre-accelerator system is shown in fig. 4. When assembled (see left insert, fig. 5) the system was held in place in a lucite mounting block by two plastic clamps (not shown). This

assembly was designed to replace the standard spectrometer source holder which fits into the source chamber from the top. All source elements were electrically insulated and connected to "feed through" electrodes in the top plate of the machined lucite mounting block. The decelerating and accelerating electrodes were made from 30/cm mesh nickel grid which has $\geq 90\%$ open area*. In initial tests lucite insulating rings were tried and were found unsatisfactory for potentials greater than 6 kV. Upon replacing these with insulating rings of Teflon no difficulty was experienced with potentials up to 12 kV. In order to limit damage caused by arcing, a test meter (6000 V, 50 μA movement) was connected in series with the high potential. This served both as a current surge limiter and also as an indicator of the steady voltage drop due to leakage current; the latter was typically only a few percent of the applied voltage.

As noted in the previous section, the sensitivity of this method is proportional to the voltage gradient V_o/L at the surface of the source foil. However, the potential V_o at the first grid must always be less than that which would repel the electrons of interest. It is thus of advantage to minimize the gap between the source foil and first grid. In our first experiments the gap was 5 mm and in later runs it was reduced to 2.6 mm. Further reductions could undoubtedly be made without difficulty.

Electrons emitted from forward-directed atoms ($\cos\theta \approx 1$) can be identified by observing them in coincidence with alpha particles emitted in the backwards direction ($\cos\theta \approx -1$). The experimental arrangement used for coincidence measurements is shown schematically in fig. 5. In order to minimize the random counting rate a short resolving time is desirable, so the Geiger electron counter was replaced by an anthracene scintillation detector.

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*Made by Buckbee Mears Corp., St. Paul, Minn.

In order to maximize light collection and prevent evaporation of this crystal, it was covered with 200 $\mu gm/cm^2$ aluminum leaf. With an applied potential of 1750 volts the noise spectrum ended at an equivalent electron energy of ~10 keV, and 25 keV electrons could be detected with high efficiency (> 70%) and an acceptably low noise background. A plastic scintillator (Naton 136) was used for detection of the alpha particles, and no signal-to-noise problem was experienced. However, the alpha pulse height spectrum was very broad, presumably because of the large energy loss and straggling caused by traversal of the relatively thick source mounting foil (~3.5 mg/cm^2 Al). It might be noted that the use of photomultipliers requires the use of magnetic shields, which is undesirable in an iron-free spectrometer. However, it was found experimentally that the focusing properties of this instrument were not unduly affected for the present purpose.

The amplifiers, single channel analyzers, and "fast-slow" coincidence circuits shown as boxes in fig. 5, are part of a commercially available system* based on a design by Chase[9]). The bias settings of the two electron single channel analyzers were matched by use of a pulser and the fast discriminators of all four analyzers set for an output pulse width of $\tau_o \approx 60$ nanoseconds. This resulted in coincidence resolving times of $2\tau_o \approx 0.12$ μsec. in both circuits. In some experiments the right hand circuit was operated in the delayed position (0.4 μsec) so that scaler No. 5 recorded the random coincidence rate. In other experiments the two electron pulse height analyzers had different settings and both coincidence circuits were "prompt," i.e., recorded the true-plus-

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*The model 901A amplifiers and model 801 coincidence unit (containing the single channel analyzers) were manufactured by Cosmic Radiation Labs., Inc., Bellport, N. Y.

random rates. The settings for the alpha channels were not critical and the counting rate in one or both was periodically monitered manually in scaler No. 1; this rate was typically $\sim 1 \times 10^4$ counts/sec.

5. Results and Analysis

In discussing our experimental results we shall distinguish between the data taken with and without the use of 180° alpha–electron coincidences. In order to interpret the results in the simple manner outlined in Section 2, where the assumption of forward directed recoils ($\cos\theta = 1$) is made, it is evidently necessary to define the recoil direction by coincidence selection; i.e., to record in the spectrometer only those electrons coincident with alpha particles emitted in the backward direction. However, in some situations where this method might be applied it may be inconvenient or impractical to record coincidences with the heavy particles, therefore we also show here results obtained with use only of the spectrometer and pre–accelerator system, in which case electrons emitted from isotropically recoiling atoms are detected in the spectrometer. These results indicate that even without the use of coincidences, useful measurements can be made.

Figure 6 shows the L_{II} and L_{III} conversion lines of the 42.8 keV transition in Pu^{240} as scanned without coincidences, with use of several different values of the electric field, and with field off. One sees qualitatively the expected result, that the Doppler shoulder is extended to momenta above $p_o\left[1 + v_r/v_e\right]$ by the electric field, and also that the effect is more pronounced with the higher field. In figs. 7 and 8 we show the data for the L_{III} line, taken with fields of 11,450 and 39,400 volts/cm., respectively, plotted on semi-log scales after background subtraction. In each case the

slope of the high energy side of the line above $p_o\left[1 + v_r/v_e\right]$ is approximately exponential, although in the latter case some curvature is evident.

These data cannot be analyzed simply by means of eq. (4) because the recoils are emitted isotropically and thus the forward component of velocity depends on the recoil direction. The recoils emitted at large θ angles travel a shorter perpendicular distance in a given time and from these components the apparent slope or "half-life" would be shorter than the correct value, i.e., that obtained from the recoils emitted in the forward direction. Thus the resulting curve is expected not to be an exponential but rather it should asymptotically approach the correct slope. Nonetheless it is possible to analyze the isotropic recoil data in a quantitative manner, and we shall return to this analysis after discussing the coincidence results.

In the coincidence experiments, the L_{III} line was selected for scanning in the spectrometer because its scintillation pulses could be separated more easily from the rising "noise" background than those of the lower energy L_{II} line. Scans were made without electric field and with applied voltages of 3, 4, 6, and 10 kV. Nominal source-to-grid distances of 5 and 2.6 mm were used.

Figure 9 shows the coincidence results obtained with applied fields 11,450 and 18,700 volts/cm, as well as with field off, and fig. 10 shows the data for 39,400 volts/cm. For comparison, the "singles" electron lines, recorded simultaneously, are also shown.

In calculating the half-life from the coincidence data we must include a small $\cos\theta$ correction to v_r because of the finite sizes of the alpha detector and source and because of a slight misalignment of the alpha detector. From the geometries involved, this correction is estimated to be ~5%, and the corrected value becomes $\bar{v}_r = 0.95 \cdot v_r = 2.65 \times 10^7$ cm/sec. The results are shown in Table Ia.

Returning now to the non-coincidence (isotropic recoil distribution) data, we note from a comparison of figs. 8 and 10, taken under similar conditions of geometry and electric field, that the slope of the ϵ curve is steeper in the case of isotropic recoils than for forward directed recoils. That this is expected has already been mentioned qualitatively. For a quantitative description of the isotropic recoil data, we proceed as follows: Recall that, in the idealized case, the momentum distribution of Doppler-shifted electrons (with field off) is constant from p_o to $p_o + \Delta p$. Since $\Delta p/p \ll 1$ the energy distribution may also be considered essentially constant from E_o to $E_o + \Delta E$ and it therefore also appears as a rectangular Doppler shoulder. For convenience we define the electron energy above E_o in units of the shoulder width ΔE, that is $\epsilon = (E - E_o)/\Delta E$. In these units, the Doppler shift energy, X, of electrons arising from atoms with a forward velocity component $v = v_r \cdot \cos\theta$ is given by $X = \cos\theta$.

On application of the electric field, V_o/L, the intensity distribution $I(\epsilon)$ from an element dX at X becomes an exponential starting at $\epsilon = X$, and has the half-value

$$\epsilon_{1/2} = \epsilon_{1/2}(\max) \cdot X \qquad (5)$$

where

$$\epsilon_{1/2}(\max) = T_{1/2} \cdot v_r \cdot (V_o/L) \cdot \Delta E^{-1} \qquad (6)$$

If the spectrometer is set to record electrons in an interval $\delta\epsilon$ at ϵ the observed intensity contribution from one element dX (for $X < \epsilon$) is given by

$$I(\epsilon)\,\delta\epsilon = \delta\epsilon \cdot I_o(X) \cdot e^{-\lambda\,(\epsilon - X)} \qquad (7)$$

where $\lambda = 0.693/\epsilon_{1/2}$,

and $I_o(X) = \lambda dX$.*

The total contribution from all elements is thus

$$I(\epsilon)\,\delta\epsilon = \delta\epsilon \int_o^k \frac{0.693}{\epsilon_{1/2}(\max)} \cdot \frac{e}{X}^{-\frac{0.693\,(\epsilon - X)}{\epsilon_{1/2}(\max) \cdot X}} dX$$

$$= \delta\epsilon \cdot \frac{0.693}{\epsilon_{1/2}(\max)} \cdot e^{\frac{0.693}{\epsilon_{1/2}(\max)}} \left[- \mathrm{Ei}\left(- \frac{0.693\,\epsilon}{\epsilon_{1/2}(\max)} \cdot \frac{1}{k} \right) \right] \qquad (8)$$

where Ei (-x) is the exponential integral, $\int_{-\infty}^{-X} \frac{e^t}{t}\,dt$, and the integration limits are: $k = 1$ for $\epsilon \geqslant 1$, and $k = \epsilon$ for $\epsilon < 1$.

Using this expression and tabulated values of the exponential integral[10], we have computed theoretical intensity distributions, $I(\epsilon)$, for several values of the parameter $\epsilon_{1/2}(\max)$. These are displayed in fig. 11. It should be emphasized that these curves describe only the effect of the electric field on the electrons in the Doppler shoulder.

One can deduce the half-life from a measure of the slope of the observed distribution at any energy value above the shoulder ($\epsilon > 1$). The observed half-value $\epsilon_{1/2}(\epsilon)$ may be related to the asymptotic value $\epsilon_{1/2}(\max)$ with use of the

*The integrated intensity, $\int_X^\infty I_o(X)\, e^{-\lambda\,(\epsilon - X)}\, d\epsilon = I_o(X)\, \lambda^{-1}$, must equal the original intensity from element dx; therefore, $I_o(X) = \lambda dX$.

curves given in the inset of fig. 11, which have been calculated for several convenient values of ϵ. From the value of $\epsilon_{1/2}(\max)$ so obtained, the half-life may be calculated directly, with eq. (4).

The non-coincidence data have been analyzed in this manner, and the results are shown in Table Ib. The important fact to note from a comparison of Table Ia and Ib is that for data taken under the same conditions there is very little deviation between the results taken with and without the use of 180° alpha-electron coincidences, which is an indication of the validity of the analytical treatment of the non-coincidence data.

To assess the overall accuracy of the data it is helpful to examine the sources of error in determining each of the three quantities in eq. (4). With respect to the first quantity, $\epsilon_{1/2}$, there is relatively little error since the statistical accuracy of these "decay curves" is quite good. The value of the electric field, V_o/L, contains errors both in the measurement of the high voltage and of the source-to-grid distance. In the earlier runs before it was certain that the data could be treated quantitatively careful measurements were not made of the voltage applied to the grid, and this was the case with the run at 11,450 volts/cm (6 kV). The source-to-grid distance (~5 mm) was known to better than 5%, and we attach an overall error of ±10% to this value of V_o/L. In the later runs at 10 kV a "field emission" line (electrons emitted from the grid and accelerated through the full applied potential) was observed in the spectrometer and the energy of this line defined the voltage to better than 1%. However, in these later runs the source-to-grid distance had been shortened to 2.6 mm, and the uncertainty in this distance is thought to be about 10%. Therefore, in all cases the value of V_o/L has an uncertainly of about 10%. Since this study was initiated in order to test the feasibility of the method rather than to make precision

measurements, great care was not taken in the construction of the grid system, and it is certain that with some effort the accuracy in determining V_o/L can be substantially increased.

From consideration only of the above errors, we would quote a value of the half life $T_{1/2} = (1.6 \pm 0.2) \times 10^{-10}$ seconds. This value is reasonably close to that measured electronically by Bell, Bjørnholm, and Severiens $(1.73 \pm 0.15) \times 10^{-10}$ seconds[11]). However, another source of error in our measurements, not yet discussed, is more difficult to evaluate. This is associated with the determination of v_r, the recoil atom velocity. Because of the finite thickness of the Cm^{244} source, some slowing-down of the recoiling atoms occurs so that they emerge not with the calculated velocity v_r but with a somewhat reduced average velocity $\bar{v}_r$. Evidence that this is so may be seen in fig. 9, where we note that the energy shift of the "Doppler peak" (electrons observed in coincidence with backward-directed alpha particles, with no electric field) is ~30% less than expected on the basis of full forward recoil velocity. Thus $\bar{v}_r$ may in this case be as much as 30% lower than v_r and therefore our measured half-life value might be higher by as much as 30%. It would be desirable to measure $\bar{v}_r$ directly, and this can probably be done by a time-of-flight technique.

6. Discussion

The experiments described here have shown that a nuclear lifetime of 10^{-10} seconds following alpha decay is easily measurable by the electrostatic technique in conjunction with a high-resolution beta-ray spectrometer. It is of interest to speculate on the potentialities of this method under various experimental situations. In order to make the discussion quantitative we

have prepared a set of illustrative diagrams, figs. 12 to 15. Figure 12 shows the maximum electron Doppler shift for some typical reactions. Note that in all cases the Doppler shift is $\geq 0.1\%$ in momentum and thus is clearly visible with spectrometer resolutions of $\leq 0.1\%$. Of interest in the application of this method is the additional shift in electron momentum produced by the electrostatic grid system. In fig. 13 we show calculated values of this "electrostatic shift" for two reactions and several lifetimes.

To assess the range of applicability it is useful to compute the electrostatic shifts one would expect for the transition multipolarities most frequently encountered. Using the single-particle lifetime formula[12] corrected for internal conversion (see fig. 14) we obtain the graphs shown in fig. 15. The left-hand set is representative of the typical situation following alpha decay in heavy elements. The right-hand set illustrates the case of a rare-earth target bombarded by heavy ions.

With spectrometers of the iron-free type one can easily determine line momenta with a precision of $< 0.01\%$. Under favorable conditions (thin source, good statistics) one might then expect to observe "electrostatic shifts" as small as $\sim 0.01\%$ in momentum. From fig. 13 we see that this would correspond to lifetimes of $\sim 3 \times 10^{-12}$ seconds for low energy transitions following alpha decay and $\sim 3 \times 10^{-13}$ seconds for a typical heavy-ion reaction. One can hope to improve the limit of sensitivity with higher electric fields (closer grid spacing) and particularly for more energetic conversion electrons larger decelerating voltages, V_o, can be used. The limit of 0.01% in momentum is shown in each portion of fig. 15 as a horizontal straight line. We see that for heavy-ion reactions it should be possible to measure lifetimes of magnetic dipole transitions up to ~200 keV in energy if they proceed at the single-particle rate and correspondingly higher in energy if they are retarded. The situation is quite favorable for E2 transitions.

As was noted earlier this method in fact measures the distance which the recoiling atoms travel before emitting conversion electrons. For example, a rare-earth atom recoiling from absorption of an 80 MeV O^{16} nucleus has a velocity of 3×10^{8} cm/sec and hence travels 0.03 mm in 10^{-11} seconds. This distance is small compared to the source dimensions used in this work, 1 mm × 10 mm. However, for lifetimes $\geq 10^{-9}$ seconds the distances are ≥ 3 mm which would result in a serious smearing of apparent source dimensions in the case of recoil distributions with a large angular spread (e.g. Coulomb excitation). Fortunately, for most heavy-ion reactions the recoil distribution pattern is strongly peaked in the forward direction and one might expect to be able to work with lifetimes as long as 10^{-9} seconds. In the case of longer lifetimes it might be possible to analyze the change in line shape to give a direct measure of half life if the emitted conversion electrons are observed with a high-resolution spectrometer at right angles to the recoil direction. In the above example 3 mm travel in the spectrometer radial direction corresponds to an apparent shift of 0.15% in momentum, in a 50 cm radius $\pi\sqrt{2}$ spectrometer. With appropriate geometry one would thus expect to observe an exponential high energy tail on the conversion line.

A crucial requirement for the success of the electrostatic method is that the sources or targets be thin. We note that the line shapes from the Cm^{244} source showed evidence of appreciable energy degradation of the recoiling Pu^{240} atoms in the source material. The mean surface density of this source was > 10 $\mu g/cm^2$. For the case of heavy-ion reactions one expects to be able to use somewhat thicker targets (sources); for example a mass-150 atom recoiling from absorption of an 80 MeV O^{16} nucleus has a kinetic energy of ~10 MeV and a mean range of ~750 $\mu g/cm^2$ in aluminum[13]). Hence one could tolerate a

target thickness of ~70 $\mu g/cm^2$ with a resulting energy spread of ~10% or velocity spread of ~5% for the emerging atoms. This would mean that (relativistic) conversion electrons emitted in the forward direction would have up to 5% less than the full Doppler shift (~1% in momentum). That is, a spread of ~0.05% in momentum would be caused by absorption. There would be little or no electron energy degredation within the 70 $\mu g/cm^2$ target since almost all electrons would be emitted after the atom had left the surface (unless the nuclear lifetime was shorter than the traversal time through the target layer, i.e. $\sim 10^{-13}$ seconds). Although it is desirable to use as thin a target as possible a lower limit is in any case set by reaction yield consideration. With an ion beam of ~2 μA (singly charged), incident on a 70 $\mu g/cm^2$ target and $\sigma \sim 0.5$ barns the predicted "source strength" is $\sim 10^8$/min, which is adequate for high-resolution electron spectroscopy.

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FOOTNOTES AND REFERENCES

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Table I. Deduction of half-life of 42.8 keV state in Pu^{240}

a. With 180° α-e coincidences.

V_o/L[a]	$\epsilon_{1/2}$	$E_{1/2}$, e.V.	$T_{1/2}$(sec)
11,450[b]	0.308	48.3	1.59×10^{-10}
18,700[c]	0.458	72.0	1.45×10^{-10}
39,500[d]	1.147	180	1.72×10^{-10}

b. Without coincidences.

V_o/L[a]	ϵ	$\epsilon_{1/2}$	F	$\epsilon_{1/2}$(max)	$E_{1/2}$(max), e.V.	$T_{1/2}$(sec)
11,450[e]	1.5	0.25	1.24	0.310	48.6	1.52×10^{-10}
39,400[f]	1.5	0.708	1.77	1.255	196	1.78×10^{-10}
	2.0	0.795	1.63	1.292	203	1.84×10^{-10}
	3.0	0.897	1.46	1.310	205	1.86×10^{-10}
	4.0	0.945	1.34	1.265	198	1.80×10^{-10}
					mean =	1.82×10^{-10}

[a] Electric field at source surface in volts/cm.

[b] Data of fig. 9, curve C

[c] Data of fig. 9, curve D

[d] Data of fig. 10, curve C

[e] Data of fig. 7, curve B

[f] Data of fig. 8, curve B

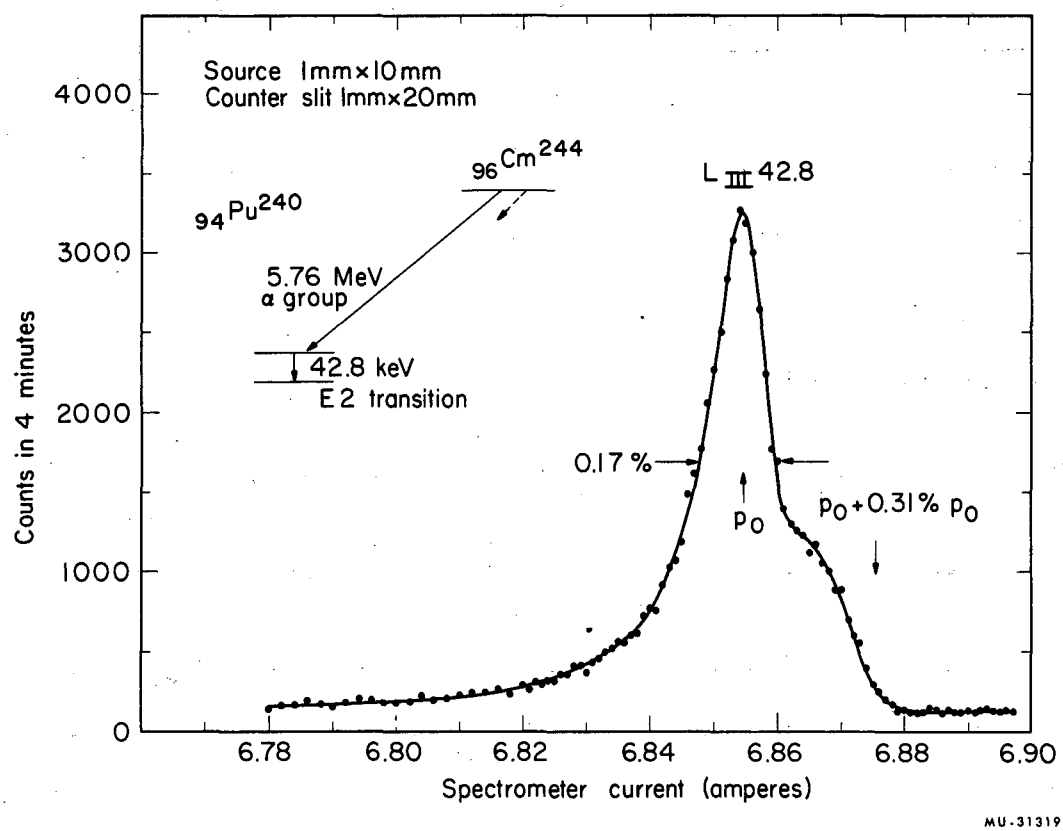

Fig. 1. Shape of the L_{III} 42.8 conversion line in Pu^{240} as observed from an uncovered source of Cm^{244} in an iron free double focusing spectrometer adjusted for an instrumental resolution of ~0.07% in momentum. The high momentum shoulder is due to electrons emitted from Pu^{240} atoms recoiling from the source surface after alpha emission. If the source were infinitely thin the shoulder would be rectangular and extend to the "half height" position indicated by the arrow labelled $p_o + 0.31\% p_o$ which is the maximum Doppler recoil effect.

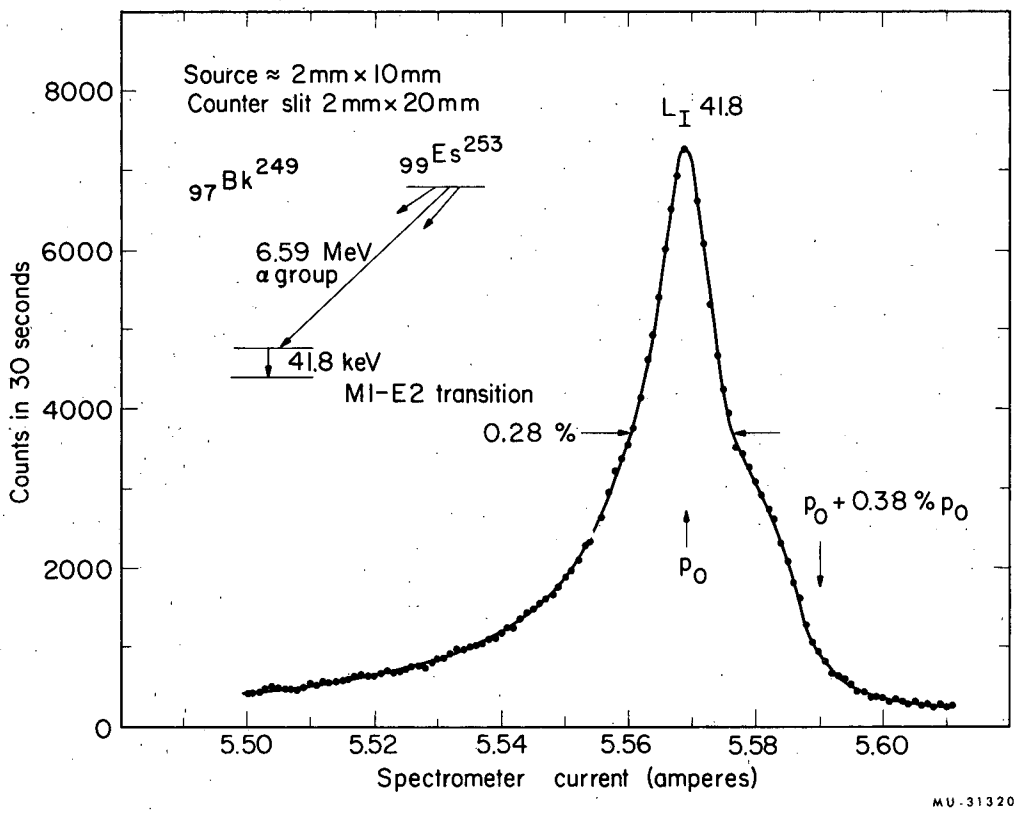

Fig. 2. Shape of the L_I 41.8 conversion electron line observed from an uncovered source of Es^{253} in an iron free double focusing spectrometer adjusted for an instrumental resolution of ~0.12% in momentum. Despite the smearing of the line shape due to source thickness, the Doppler shoulder is quite observable.

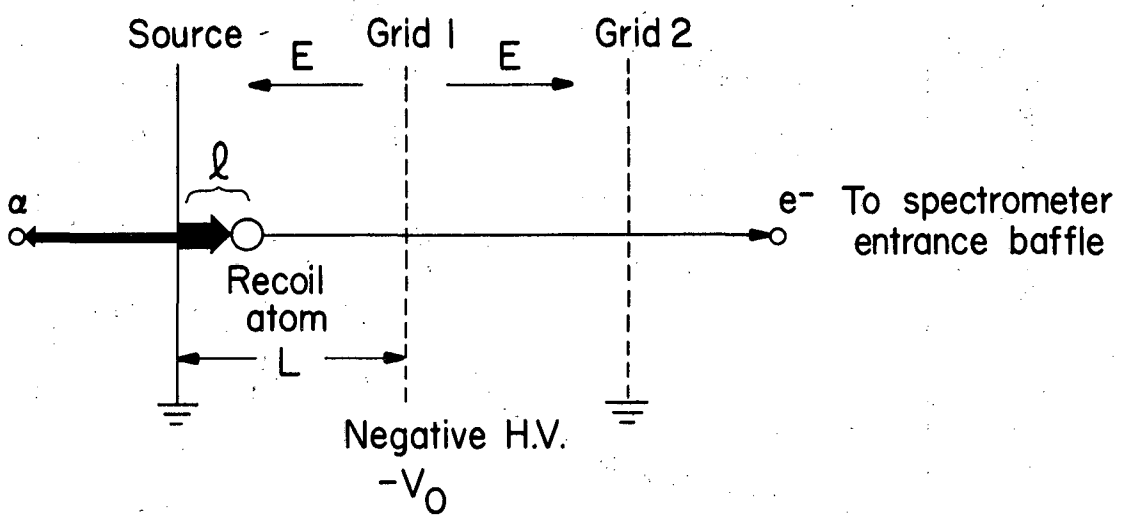

Fig. 3. Basic features of the electrostatic preaccelerator system used in this work. The maximum Doppler shift is observed when the alpha particle is emitted backwards (to the left) and the atom recoils in the direction of electron acceptance (to the right). An electron emitted from an atom at a distance, ℓ, from the source will emerge at the right with a net gain in energy of $(\ell/L)V_0$ electron volts due to the electrostatic fields.

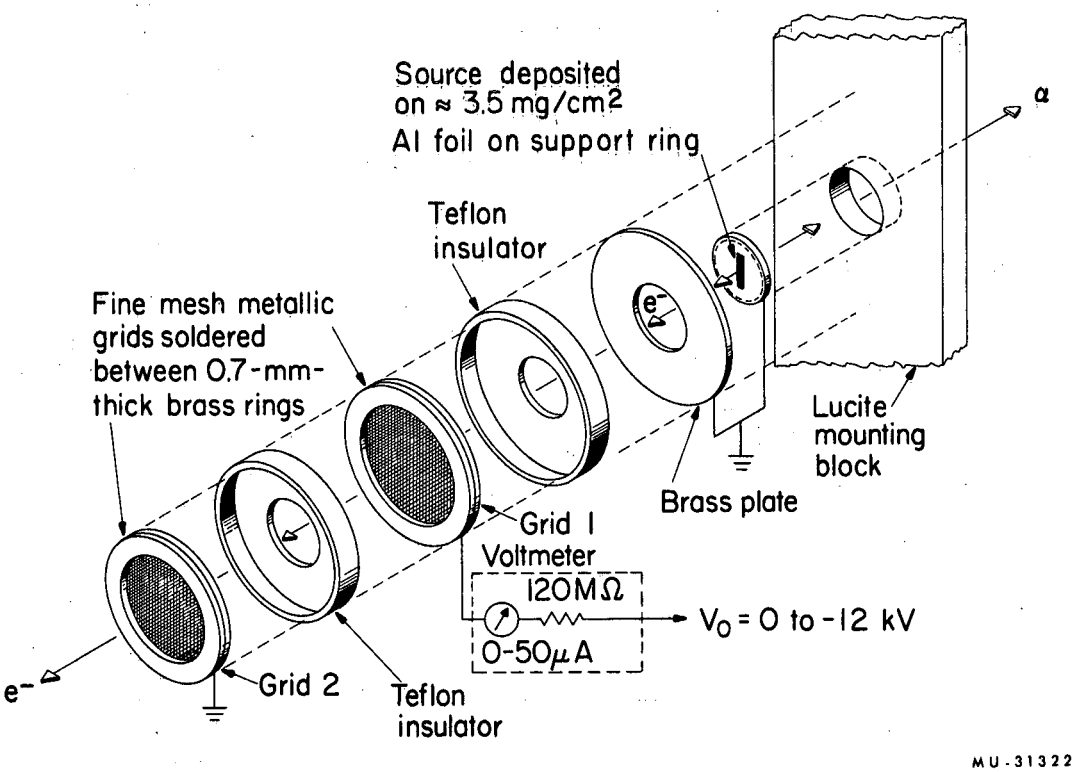

Fig. 4. Exploded view of the components of the preaccelerator system.

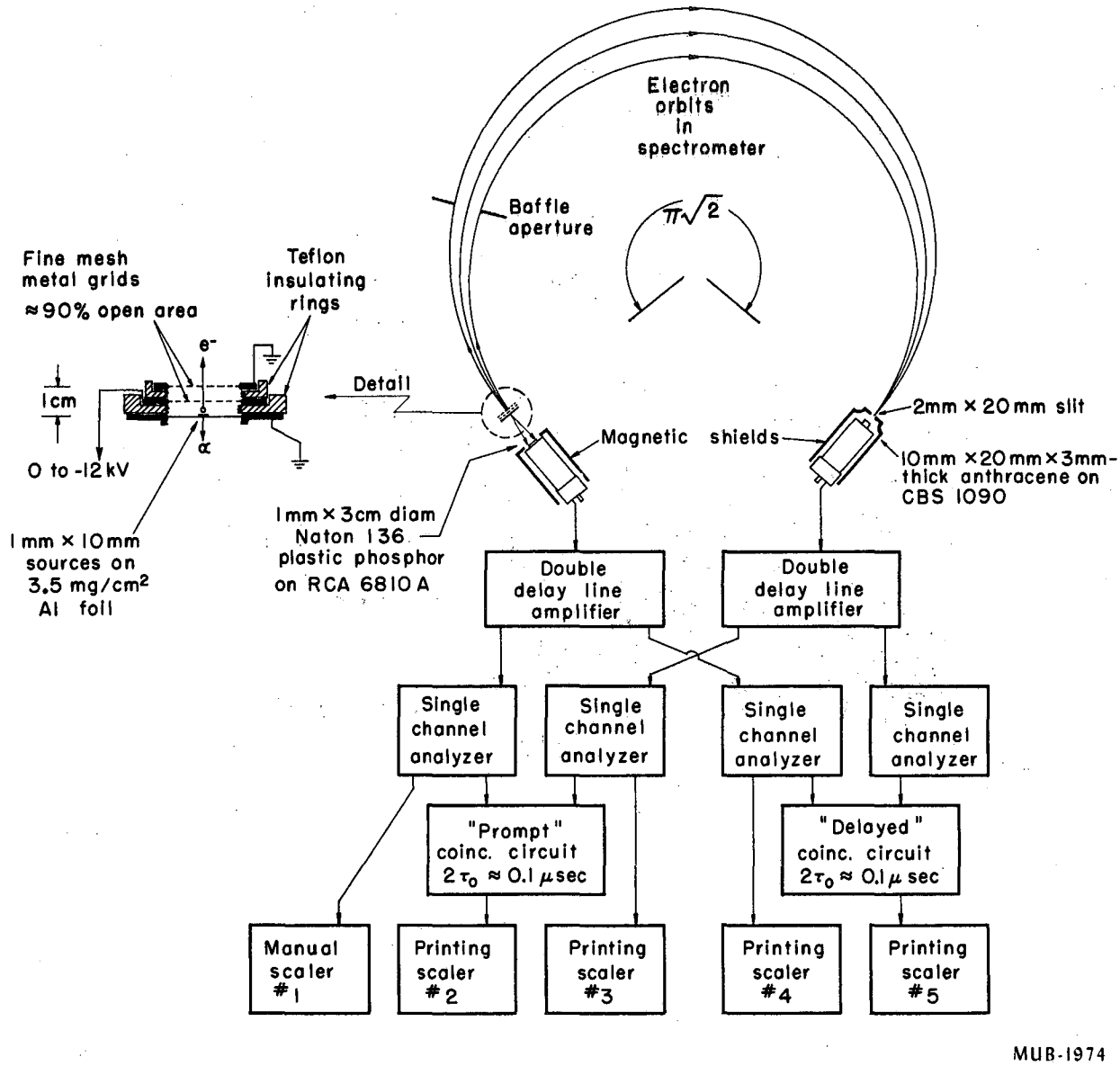

Fig. 5. Schematic block diagram of the arrangement used for measuring alpha-electron coincidences.

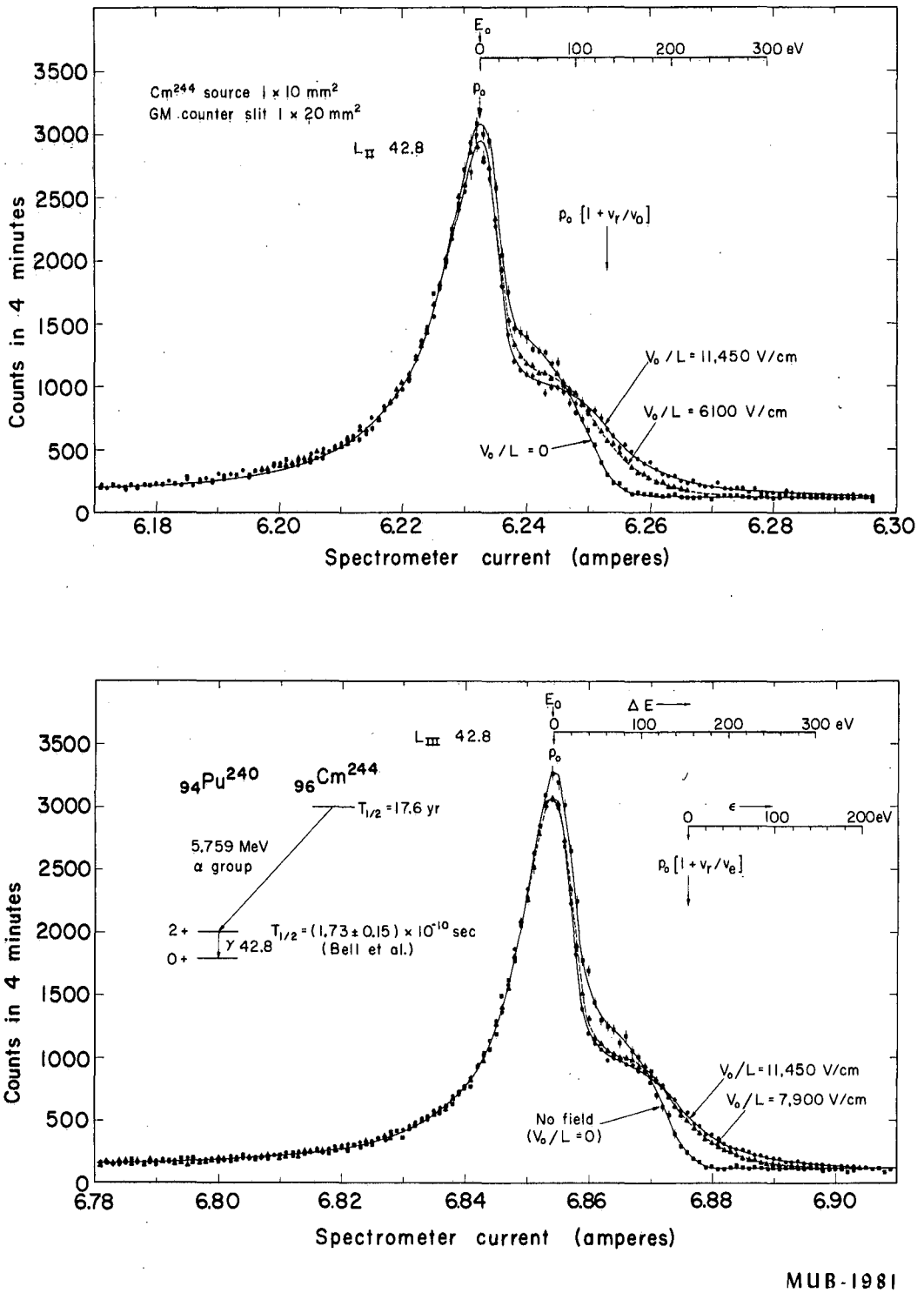

Fig. 6. Shapes of the L_{II} and L_{III} lines of the 42.8 keV E2 transition in Pu^{240} (Cm^{244} source) as measured with and without preacceleration. The shape of the Doppler shoulder is affected by the electric field whereas the main peak, due to electrons emitted from atoms at rest in the source, is not affected.

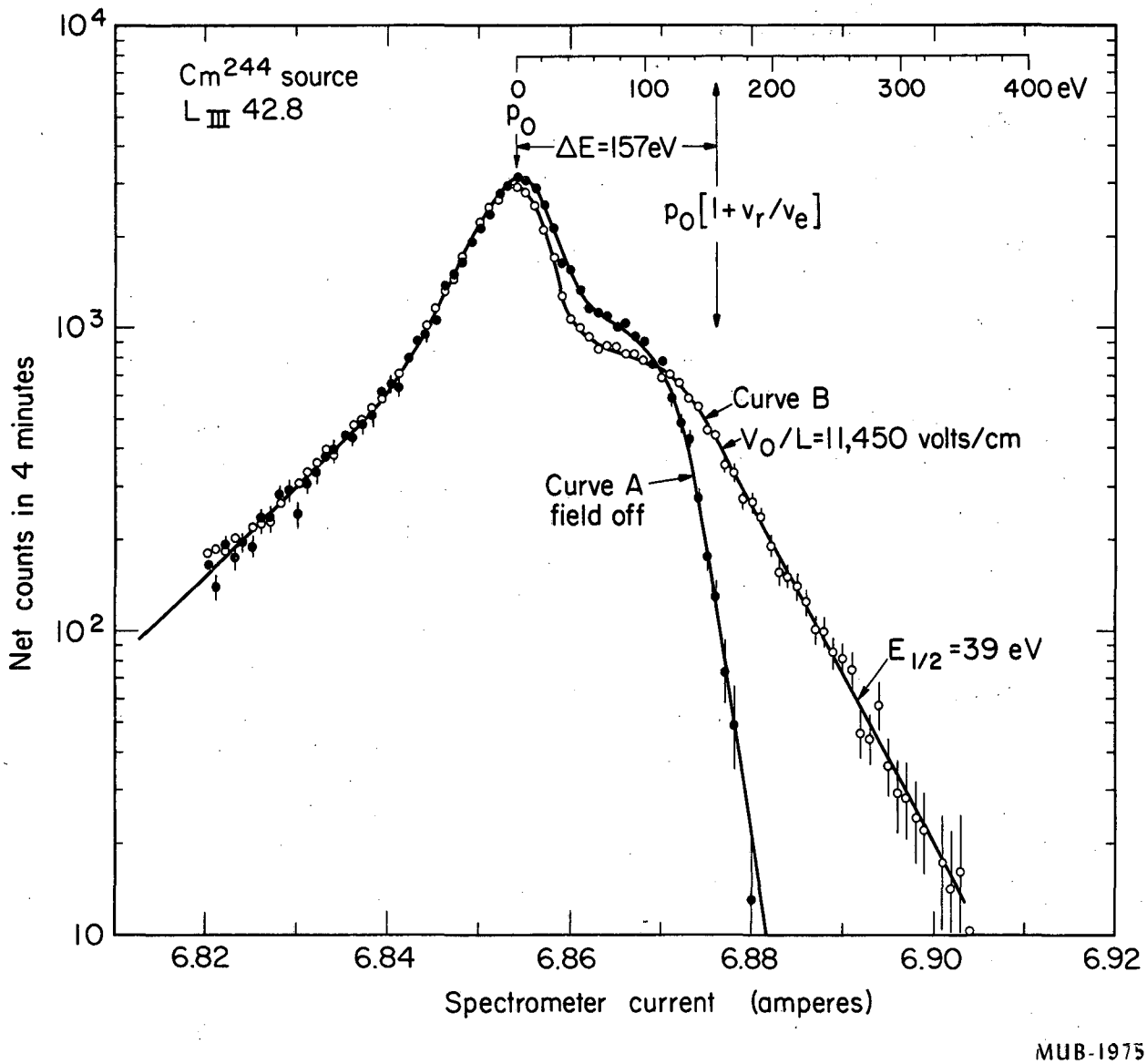

Fig. 7. A semi log plot of the data from fig. 6b after subtracting the background counting rate. With the electric field on the shape of the Doppler shoulder is altered and has an approximately exponential high energy tail.

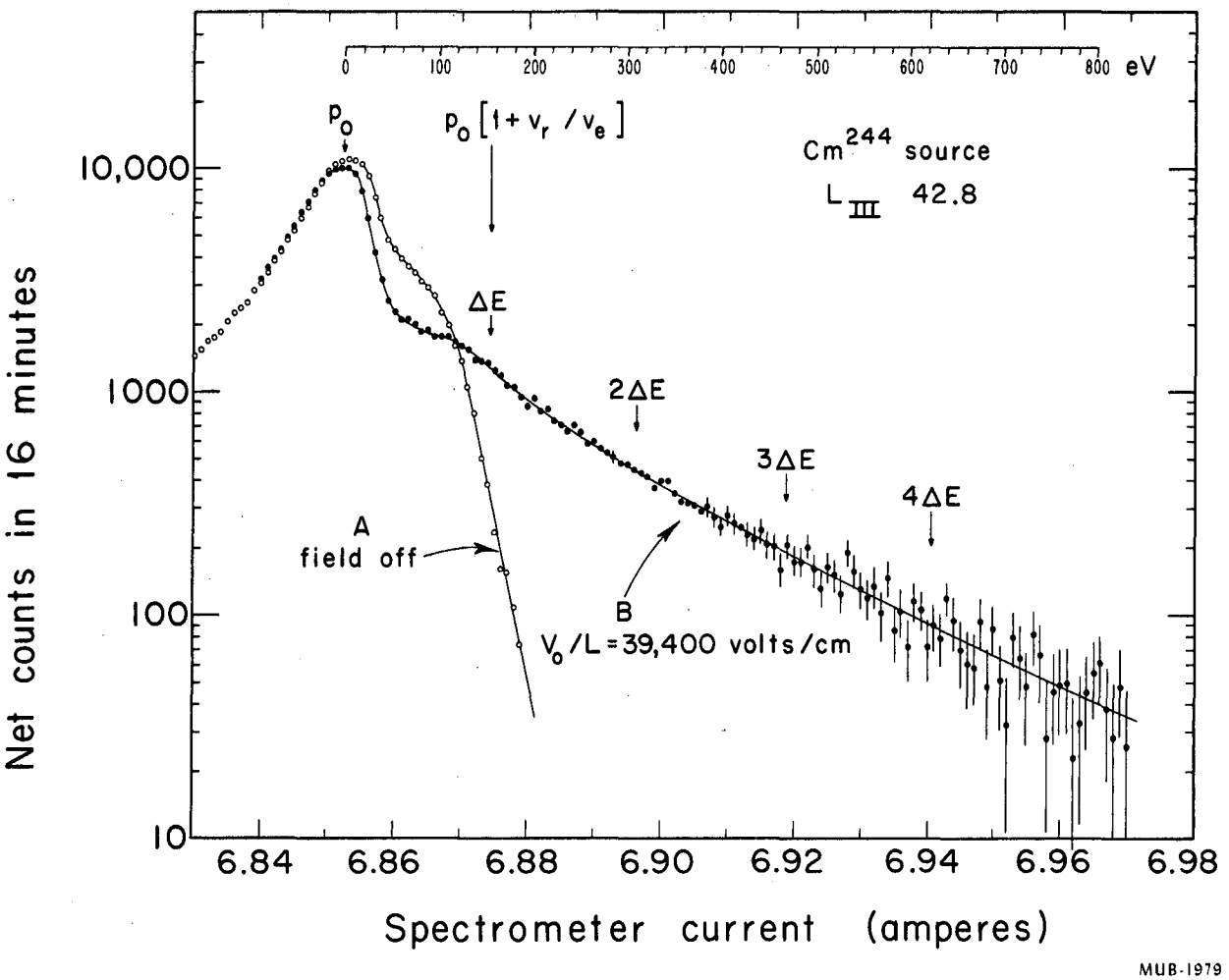

Fig. 8. Semi log plot of the L_{III} 42.8 line shape as observed with the highest electric field used in this work. This shows that the distribution of the accelerated electrons is not strictly exponential in shape but has the curvature expected theoretically (see text).

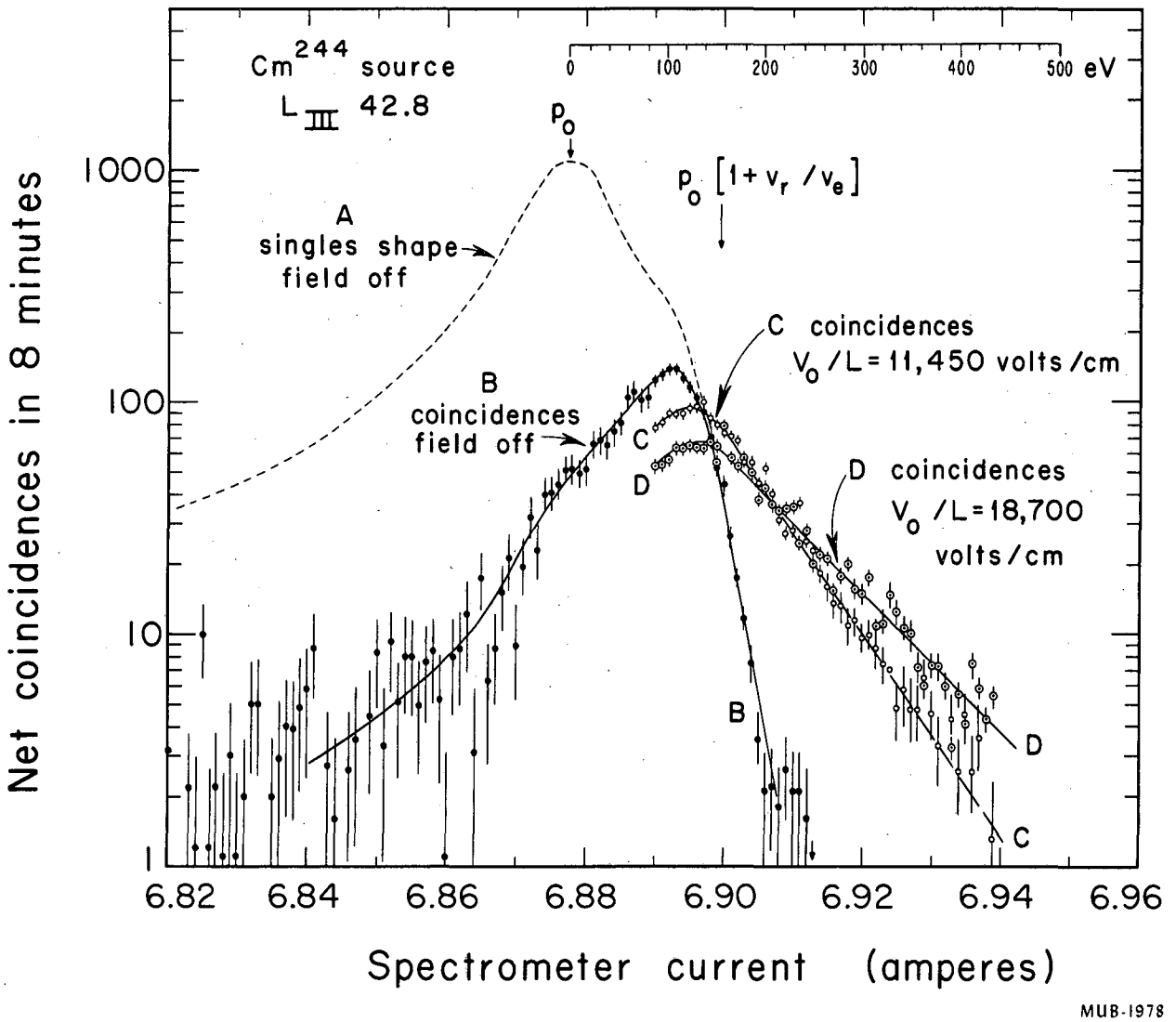

Fig. 9. L_{III} 42.8 line shapes as observed with the coincidence arrangement shown in fig. 5. Curve A is proportional to the counting rate in the electron detector after background subtraction. Similarly, the measured randon rates have been subtracted from the coincidence results before plotting the net alpha-electron coincidence rate data in curves B, C, and D.

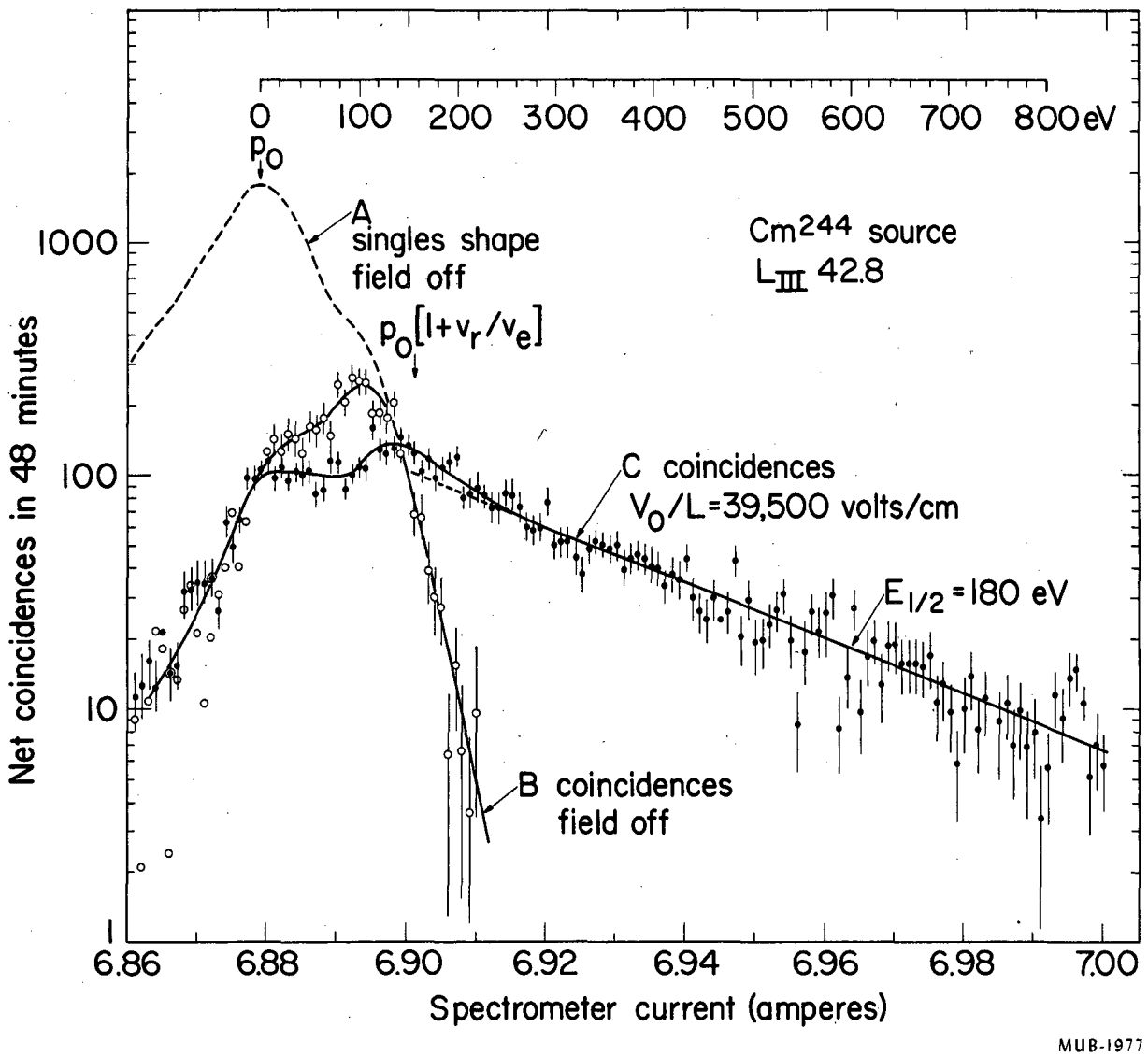

Fig. 10. L_{III} 42.8 conversion line as observed in coincidence with alpha particles at the highest preaccelerator electric field used in this work. The data were taken six weeks later than that of fig. 9 and there is some evidence of deterioration of source quality.

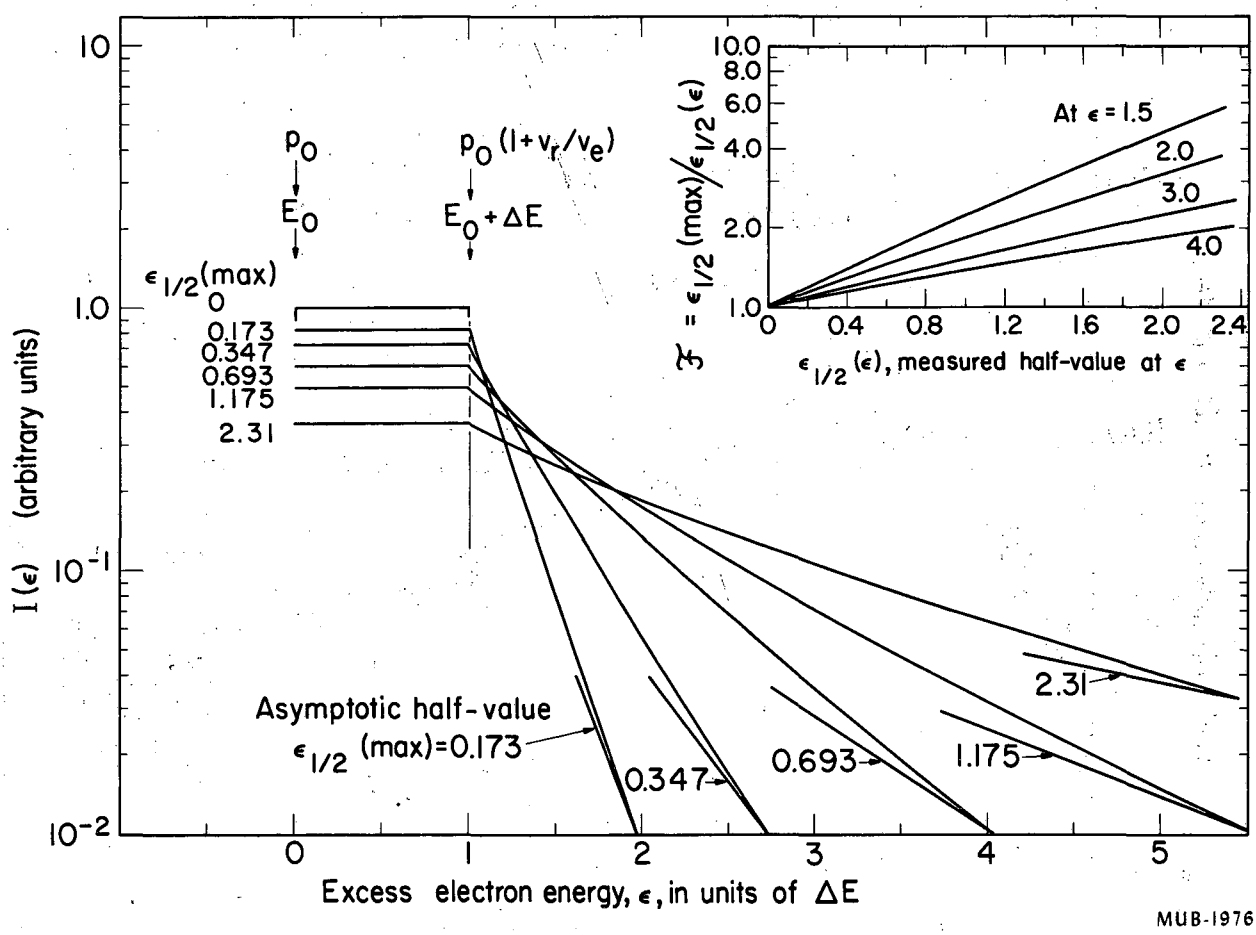

Fig. 11. Theoretical shapes of the "Doppler shift" electrons given by eq. 8 for various values of the parameter $\epsilon_{1/2}(\max)$, defined in eq. 6, which is the limiting slope at $\epsilon \to \infty$. Note that the apparent half slope $\epsilon_{1/2}$ at finite values of ϵ is shorter than $\epsilon_{1/2}(\max)$ and is not constant. The value of $\epsilon_{1/2}(\max)$ for an experimental curve may be deduced by determining the apparent half value $\epsilon_{1/2}$ at different values of ϵ and multiplying by correction factors such as those plotted in the upper right inset.

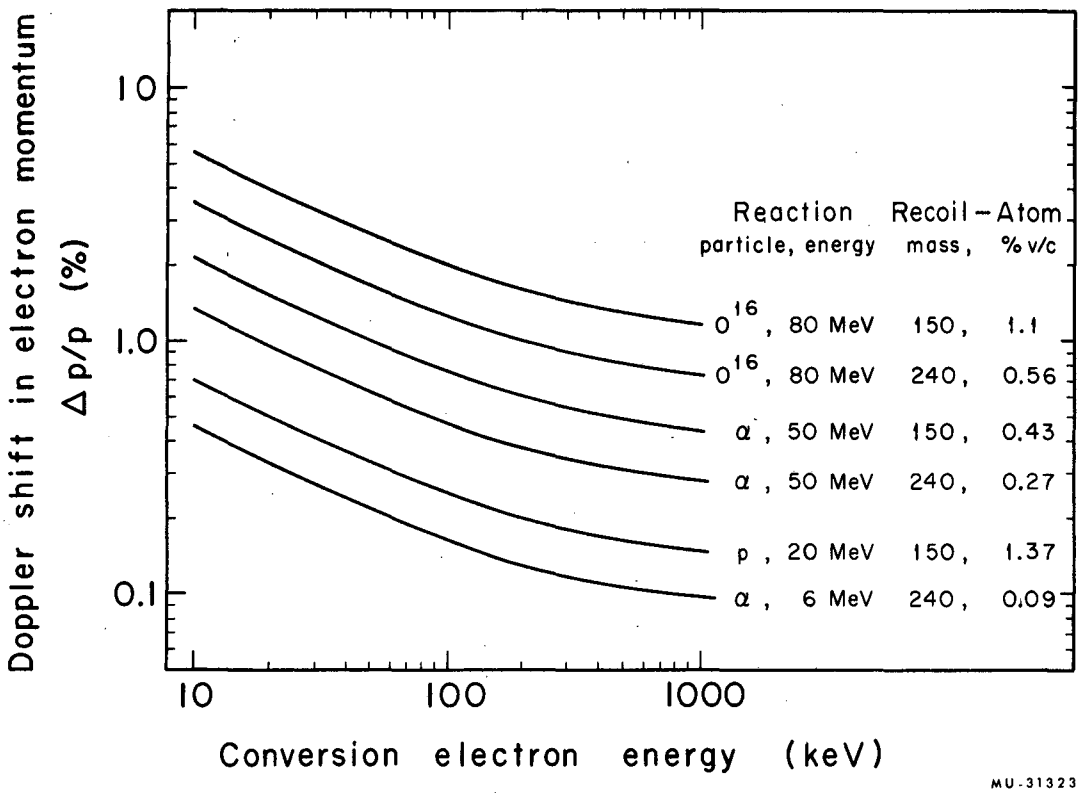

Fig. 12. Maximum Doppler shift in electron momentum for some typical nuclear reactions as a function of the conversion electron energy.

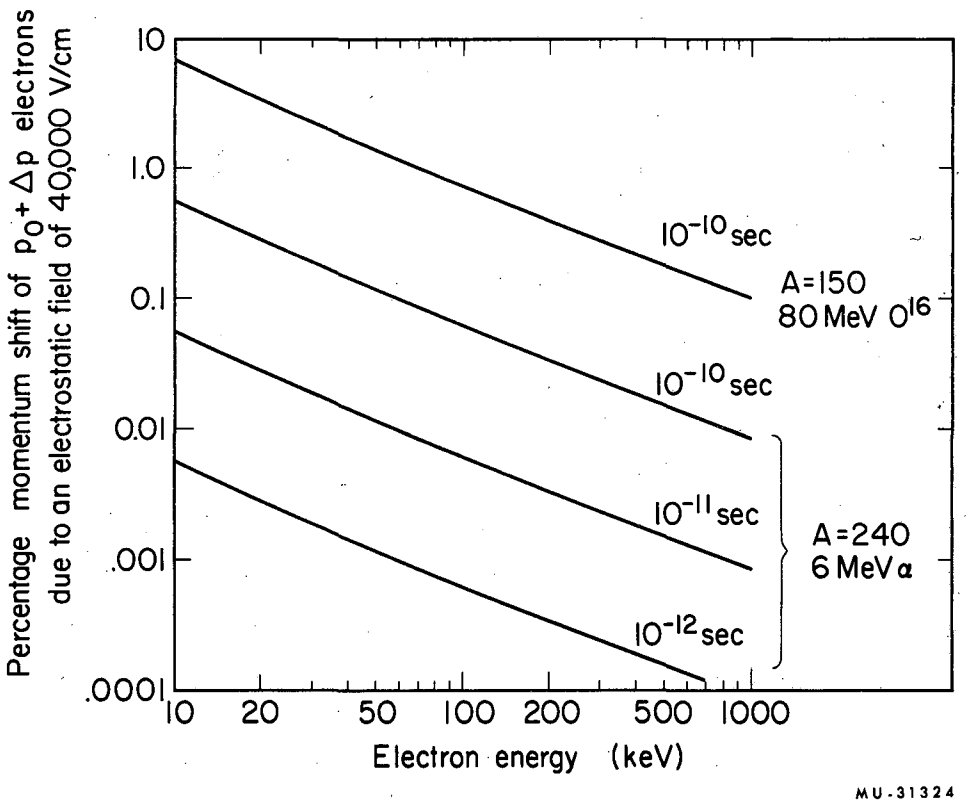

Fig. 13. Shift in the momentum of electrons with maximum Doppler energy ($\cos \theta = 1$) caused by the preaccelerator system of fig. 4 with an electric field of 40,000 volts/cm.

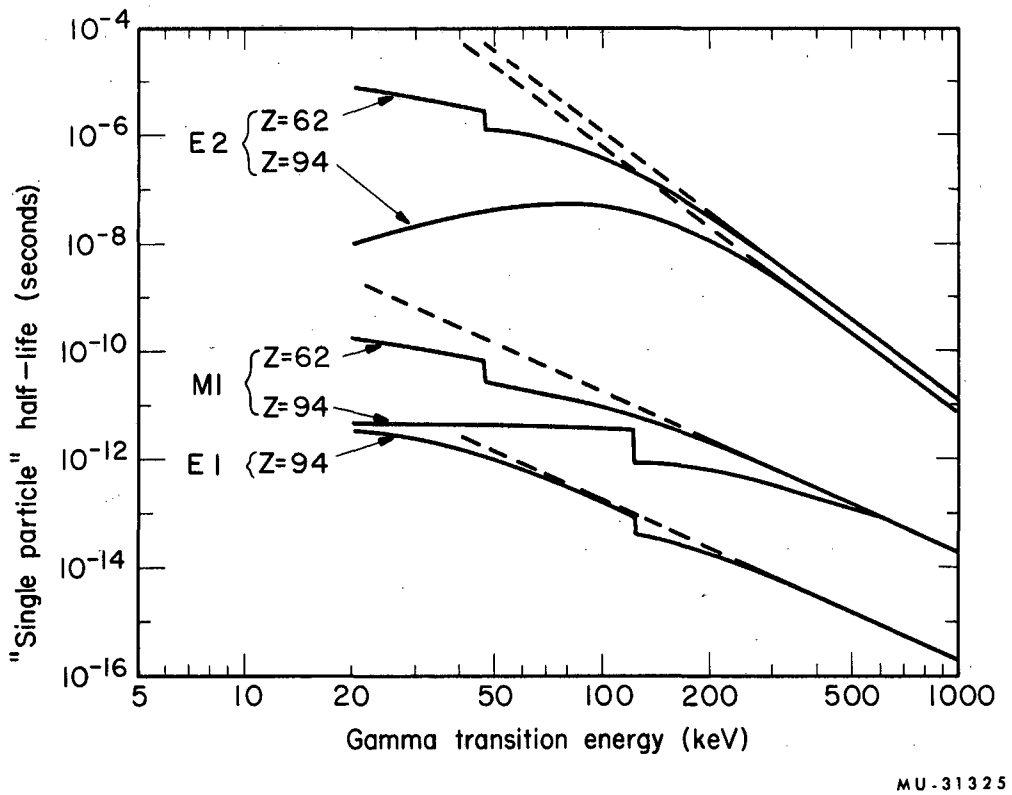

Fig. 14. Half lives of gamma transitions according to the "single particle" lifetime formula[12]) after correcting for internal conversion.

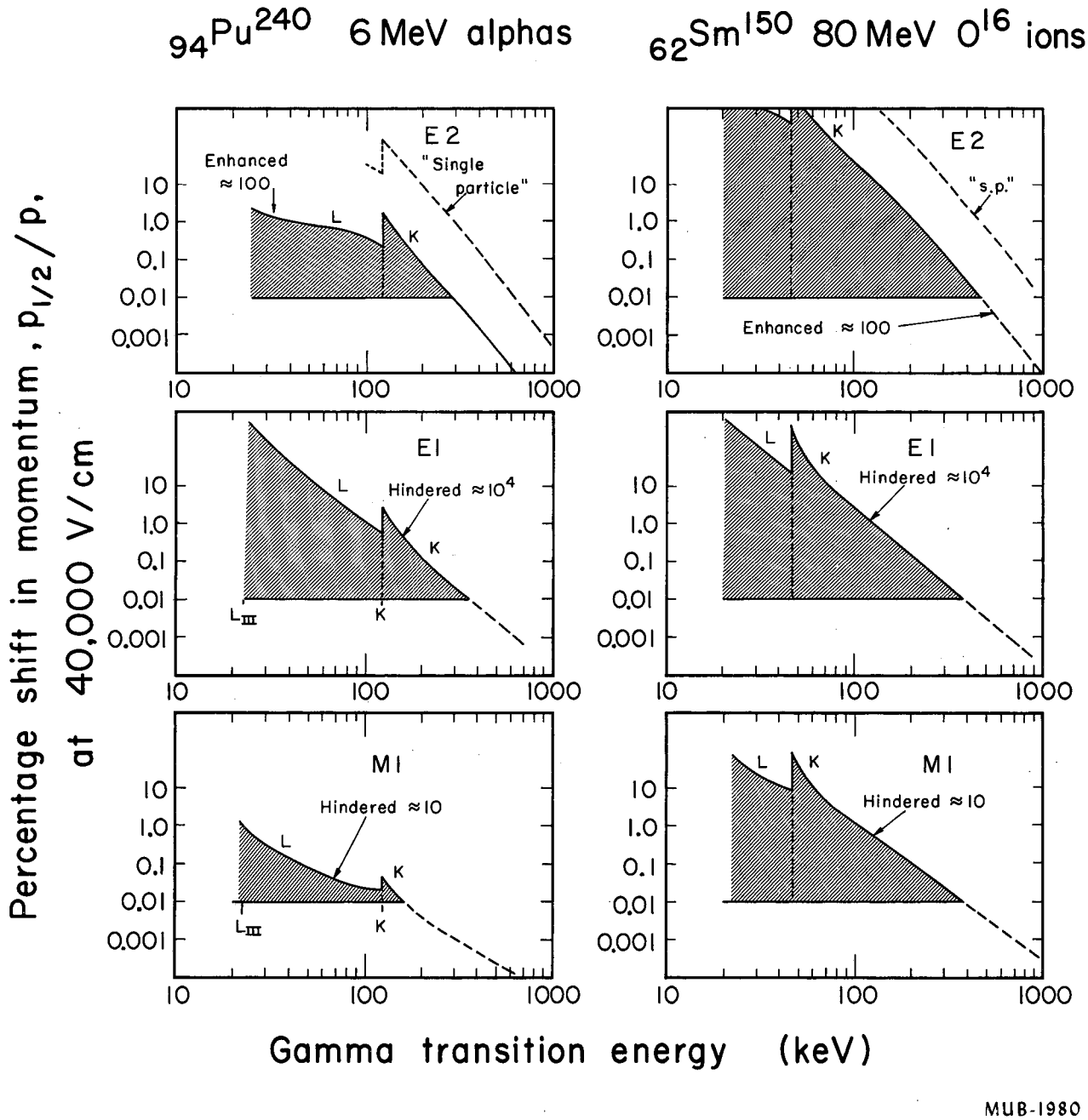

Fig. 15. Predicted half values of the electrostatic shift in momentum, $p_{1/2}/p$, with an electric field of 40,000 volts/cm for "single particle" transitions hindered or enhanced by the factors indicated. The examples on the left are representative of the situation following alpha decay of a transuranic nucleus. Those on the right are for a typical reaction involving heavy ions. If one sets 0.01% in momentum as the limit of detection (see text) then the range of applicability of this method is indicated by the shaded regions.